Conventional and Advanced Food Processing Technologies

Edited by Suvendu Bhattacharya

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Edited by

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Dedicated to

Mother
Jyotirmoyee Devi

Father
K.N. Bhattacharya

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Foreword

One of the most difficult aspects of compiling a book is to get the right mix of chapter topics. Normally, the compiling editors have the unenviable task of deciding what to put in and what to leave out, especially as many books will only have twelve to fifteen chapters. In this book, many of these issues do not exist as it is a large volume running to twenty-eight chapters and while twenty-eight topics do not exhaust the wide range of available food processing technologies, the compiling editors have come very close to making the ideal selection.

Food process technologies are many and varied, changing in popularity with changing consumption patterns and product popularity. However, a good measure of the relevance of individual unit process operations is the frequency of their occurrence in publically funded research proposals across the world. While process technologists, myself included, will often lament the lack of specific research funding for process technologies, they are nonetheless an essential delivery tool for every food research output and are to be found in many proposals. Even those well-established process technologies covered in this book can be found in ongoing research, demonstrating once again that the correct choices of chapters have been made.

Processing technology is an essential link in the food chain. Without these technologies we do not have food preservation of any sort, we do not have novel products and we have no tools with which to deliver good nutrition to the ever-increasing world population.

It is difficult to classify food processing technologies. Some authors use preservation methods, techniques for dividing raw materials into functional parts and techniques for reformulating them into finished products. This can be problematic as many process operations serve more than one of these functions. In this book, the problem is overcome by the simple use of two sections, one covering conventional or well-established processes and the other covering emerging or novel process technologies.

Section 1 on conventional processing covers all of the processing operations without which a book on process technology would be incomplete. Food preservation processes such as drying, thermal preservation, chilling and freezing are covered in separate chapters as are the combined preservation and cooking technologies of frying, baking and roasting. Not only is baking covered in its own technology-specific chapter but there is also a full chapter devoted to the critical associated process of dough handling and processing. Food deconstruction and reconstruction techniques are well covered in chapters on size reduction, extrusion, extraction, instantizing/agglomeration and gelling. Indeed, this latter and increasingly important technology is seldom covered in food process technology books and is to be welcomed here.

In most books, the above-mentioned topics would complete the conventional processing section. However, there are further gems of information to be found in this book with chapters on micronization and encapsulation (with a subsection on the use of supercritical fluids in water removal), flavouring and coating technologies (including edible coatings), fortification and impregnation (including osmotic dehydration and vacuum impregnation) and biotransformation in food processing. Once again, it is a pleasant surprise to find such a chapter in a food process technology book as it is seldom covered in such texts. Not only is biotransformation a widely used but not well-known process (cell growth and immobilization, hydrolysis, artificial flavour and sweetener production, etc.), it is a process that will undoubtedly increase in importance over the coming years.

Section 2 covers the topic of novel or newly developing process technologies. It is very easy for a preface writer to become excited about the possibilities offered by such emerging technologies and to thereby imply to the reader that these are somehow more important than the well-established conventional technologies of Section 1. Nothing could be further from the truth. Yes, they are new, exciting and full of promise. However, I am convinced that long into the future, the 'old reliables' of heat preservation, chilling, freezing and dehydration technologies will remain the cornerstone of the food process industry right across the world. However, we should not temper our excitement at the prospect of these new technologies.

The first set of chapters in this section cover the new alternative preservation possibilities offered by the use of ultraviolet light (for disinfection, mycotoxin elimination, enzyme inactivation) and infrared preservation and processing (for drying, baking, roasting, blanching and pasteurization). There is a chapter on microwave technologies (including an in-depth consideration of dielectric properties that is also of relevance to other chapters) together with one on radio-frequency heating (its potential uses and underlying science).

Another new heating technology showing much promise, ohmic heating, has a chapter in which its underlying science and application possibilities are well covered.

Membrane processing, which, like microwave processing, could justify its place in either section of this book, has its own comprehensive chapter. Its many subforms are examined in detail. Another novel pressure-driven technology, high pressure processing, has a separate chapter covering its heat and mass transfer potentials, its role in microbial inactivation and the problems associated with its application to nonliquid products.

There are three further chapters covering ozone processing (including corona discharge and cold plasma methods, antimicrobial action and potential applications), ultrasonic processing (its science, applications and limitations) and pulsed electric fields (principles, applications and use in cell disintegration).

Nanotechnology, so promising and of such concern, is covered in a separate chapter. The scientific world still awaits the verdict on the food processing applications of such an exciting new technology.

Finally, the topic of image analysis and machine vision is covered in a chapter on its application in intelligent sorting of poultry portions.

So what's missing? I, for one, cannot find it. This is a comprehensive treatment of the current state of knowledge on food process technology and, by the extent of its coverage and the selection of the top authors in each topic, looks set to become the definitive text in its field.

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Section 1

Conventional Food Processing

1

Drying and Dehydration Processes in Food Preservation and Processing

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1.1 Introduction

Drying is the removal of a liquid from a material (usually consisting of a macromolecules matrix) and is one of the most important and oldest unit operations used for thousand years in a variety of materials, such as wood, coal, paper, biomass, wastes and foods. According to Ratti (2001), drying generally refers to the removal of moisture from a substance. In the case of food materials, the application of drying aims to reduce the mass and usually the volume of the product, which makes their transportation, storage and packaging easier and more economic, but most important is their preservation and to increase their shelf-life. This is particularly important for seasonal foods, as they become available for a much longer period after drying. As water content decreases due to drying, the rate of quality deteriorating reactions decreases as well or is even suspended, leading to a product that is microbiologically steady.

Drying provides the most diversity among food engineering unit operations as there are literally hundreds of variants actually used in drying particulate solids, pastes, continuous sheets, slurries or solutions. Each drying method

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and the specific process parameters selected can cause undesirable effects on the product, including shrinkage, case hardening, change of the porosity and porous size distribution, colour change, browning, loss of aromatic compounds, reduction of nutrient and functional molecules, and others.

The most important and widespread drying methods are discussed in the present chapter. Emphasis has been paid on the presentation of the effects of each technique on the properties (structural, nutritional, quality) of the food undergoing drying.

1.2 Drying kinetics

A convenient way to express the reduction of moisture content of a material during drying is to use a drying kinetic equation, which expresses the moisture content or moisture ratio as a function of time. Several drying equations have been presented in the literature (Estürk, 2012). The simplest of them is the exponential model or Lewis equation, which includes a constant, known as the drying constant. This is a phenomenological coefficient of heat and mass transfer. Drying kinetics replace the complex mathematical models for the description of the simultaneous heat and mass transport phenomena in the internal layers of the drying material and at the interface with the surrounding space. It is a function of the material characteristics (physical properties, dimensions) and the drying environment properties, including temperature, humidity and velocity of air, chamber pressure, microwave power, ultrasound intensity and other factors depending on the drying method(s) used. The drying constant is determined experimentally in a pilot plant dryer based on drying experiments of the examined material under different values of the drying parameters.

1.3 Different drying processes

1.3.1 Hot-air drying

Hot-air (or conventional) drying (HAD) is one of the oldest, most common and simplest drying methods for dewatering of food materials. Thus, it is frequently used to extend the shelf life of food products. It is one of the most energy-consuming food preservation processes, but its main disadvantage focuses on the drastically reduced quality of the hot-air treated foods compared to the original foodstuff. High temperatures during HAD have a great influence on colour degradation and the physical structure of the product, such as the reduction in volume, decrease in porosity (shrinkage) and increase in stickiness. This phenomenon takes place when the solid matrix of the material can no longer support its own mass. The phenomena underlying HAD outline a complex process involving simultaneous mass and energy (mainly

heat) transport in a hygroscopic and shrinking system. The solid to be dried is exposed to a continuously flowing hot stream of air or inert fluid (N_2, CO_2) where moisture evaporates as heat is transferred to the food (Ratti, 2001).

During drying, evaporation of water desiccates the solid matrix of the food material and increases the concentration of solubles in the remaining solution. Changes in pH, redox potential and solubility may affect the structure and functionality of biopolymers, while in the final stages of drying phase transitions may occur. Increased concentration of solubles can promote chemical and enzymatic reactions due to higher concentrations of reagents and catalysts. The removed water is, at least partially, replaced by air and the contact with oxygen is substantially increased (Lewicki, 2006). The mechanisms related to the water movement include capillary forces, diffusion due to concentration gradients, flow due to pressure gradients or to vaporization and condensation of water, diffusion of water vapour in the pores filled with air and diffusion on the surface.

One of the most common dryers for many applications, including air drying of food materials, is the conveyor belt dryer, which is depicted schematically in Figure 1.1. Dryers of this type usually consist of sections placed in series,

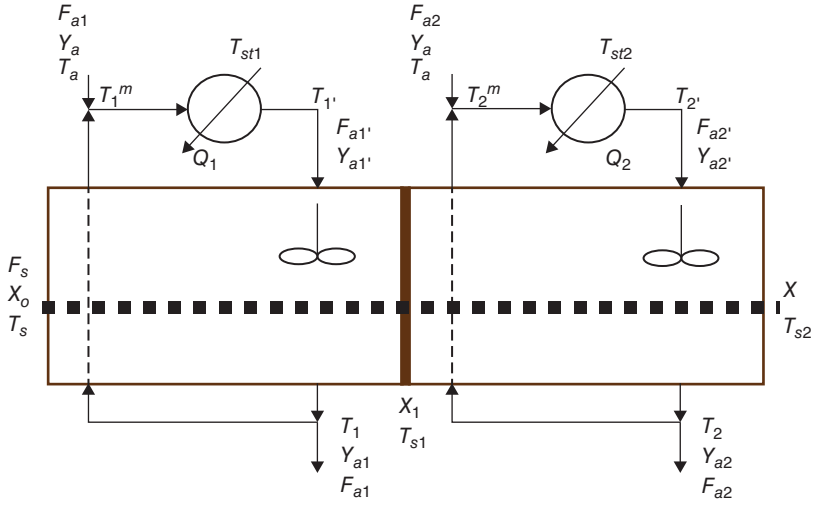

Figure 1.1 Representation of a one-section two-chamber conveyor belt dryer (F_s, dry solids feed flow rate; F_{ai}, dry air flow rate exiting chamber i after the splitter (this is equal to the fresh dry air flow rate entering chamber i, where $i = 1, 2$ in the case of the presented dryer), $F_{ai}{}'$, dry air flow rate passing through chamber i; Q_i, heat duty in chamber i; T_s, initial solids temperature; T_{si}, solids temperature exiting chamber i; T_a, ambient air temperature; T_i, air temperature exiting chamber i; T_i', initial air temperature feeding in chamber i; T_i^m, air temperature after the mixing of recirculated and fresh air in chamber i; T_{sti}, steam temperature in the exchanger of chamber i; X_0, initial solids moisture content; X_1, solids moisture content exiting chamber 1; X, final solids moisture content; Y_a, ambient absolute humidity; Y_{ai}, absolute humidity of air exiting chamber i; $Y_{ai}{}'$, absolute humidity of air feeding in chamber i)

each of which includes a certain number of chambers. The conveyor belt is common for all the chambers of a section. The properties of drying air such as temperature and velocity in each chamber can be adjusted independently from the rest of the section's chambers by means of a heat exchanger and fan installed in each chamber. Additionally, the air circulation is also independent in each chamber and through the mixing of recirculated and fresh air to the proper ratio achieves the desired properties such as that of the air humidity. The dryer presented in Figure 1.1 is a one-section two-chamber dryer.

1.3.2 Vacuum drying

Vacuum drying (VD) is an efficient technique for reducing moisture content of heat-sensitive materials that may be changed or damaged if exposed to high temperature. Characteristics of VD are the high drying rate due to the low vapour pressure in the drying environment, the low drying temperature as the boiling point of water reduces with a pressure drop, the oxygen-deficient drying environment and the reduction of energy consumption. These characteristics contribute to conservation of qualities such as colour, shape, aroma, flavour and nutritive value of the dried product (Šumić *et al.*, 2013) and induce degradation of nutritional compounds, oxidation of beneficial substances or formation of toxic compounds (Dueik, Marzullo and Bouchon, 2013). Due to molecular transport of evaporated water the process is long and can last up to 24 hours. Dry products are of very good quality but the shelf-life is dependent on the post-drying processes applied (Lewicki, 2006). VD is ideal in situations where a solvent must be recovered or when materials have to dry to very low levels of moisture.

Lee and Kim (2009) studied the drying kinetics of radish slices in a vacuum dryer at a pressure of 0.1 mPa. They observed the absence of a constant drying rate period. An increase in the drying temperature and a decrease in slice thickness caused a decrease in the drying time. The effective diffusivity varied from 6.92 to 14.59×10^{-9} m^2/s over the temperature range of 40–60 °C and followed an Arrhenius-type relationship.

Šumić *et al.* (2013) investigated VD of frozen sour cherries in order to optimize the preservation of health-beneficial phytochemicals, as well as the textural characteristics. The optimum conditions of ~54 °C and ~148 mbar were established for VD of the food material considering the maximum amount of total phenolics content, vitamin C, anthocyanin and maximum antioxidant activity and the minimum total colour change, a_w value and firmness of the product. Under optimal conditions, the value of the following quality indicators of dried sour cherry was predicted: total phenolics was 744 mg CAE (chlorogenic acid equivalents)/100 g dry weight (d.w.), vitamin C 1.44 mg/100 g d.w., anthocyanin content 125 mg/100 g d.w., antioxidant activity IC50 3.23 mg/ml, total solids 70.72%, water activity a_w value 0.65, total colour change 52.61 and firmness 3395.4 g.

1.3.3 Microwave drying

Microwave drying (MWD) results in the dewatering of a food material by heating it in a microwave oven using microwave energy, which is an electromagnetic radiation in the frequency range between 3 MHz and 30 000 GHz. The main factors affecting this method include sample mass, microwave power level and heating duration. Microwave heating of dielectric materials is governed by dipole rotation and ionic polarization. When a moist sample is exposed to microwave radiation, molecules such as H_2O carrying dipolar electrical charges rotate as they attempt to align their dipoles with the rapidly changing electric field. The resultant friction creates heat, which is transferred to neighbouring molecules. The internal temperature of a moist and microwave heated sample may reach the boiling point of water and the free moisture evaporates inside the product, causing a vapour pressure gradient that expels moisture from the sample. The internal temperature remains at boiling point until all free moisture is evaporated, followed by a rapid increase, which causes losses of volatiles, chemical reactions and eventual charring. One of the most important advantages of MWD is the reduction of drying time as heat is generated internally, resulting in a high rate of moisture removal. However, MWD possesses a few difficulties during application; these are uneven heating and underdrying or charring.

The food industry is now a major user of microwave energy, especially in the drying of pasta and post-baking of biscuits. The use of large-scale microwave processes is increasing and recent improvements in the design of high-powered microwave ovens has reduced equipment manufacturing costs. The operational cost is lower because energy is not consumed in heating the walls of the apparatus or the environment (Vadivambal and Jayas, 2007). A drawback with microwave heating is that there is no common method to monitor or control the electromagnetic field distribution and its effect after the microwave is switched on.

MWD affects most of the product properties. It has shown positive ratings for drying rate, flexibility, colour, flavour, nutritional value, microbial stability, enzyme inactivation, rehydration capacity, crispiness and a fresh-like appearance. The rehydration characteristics of microwave dried products are expected to be better as the outward flux of escaping vapour during drying contributes to the prevention of structure collapse. The quantum energy of microwaves is quite low and does not cause extensive chemical changes and thus helps in the retention of nutrient activity (Vadivambal and Jayas, 2007).

1.3.4 Freeze drying

The food material must be frozen and then subjected to dewatering by ice sublimation under very low pressure to conduct vacuum freeze drying, known simply as freeze drying (FD). FD is the result of three discrete stages:

1. Freezing stage. Initially, the product has to be frozen to achieve a solid structure that avoids collapse while the drying process is realized by sublimation. This stage has a great influence on the whole process because it sets the structure of the ice crystals (shape and size), which ultimately affects the heat and mass transfer rates. Attention needs to be devoted to the control of the uniformity of the cooling gas temperature (Hottot, Vessot and Andrieu, 2004).
2. First drying (sublimation) stage. Sublimation (solid ice transforms to water vapour without the conversion into liquid water) requires a large amount of energy (~2800 kJ/kg of ice). The heating of the frozen material generates a sublimation front that advances gradually inside the frozen solid and its temperature is practically constant. Mass transfer occurs by migration of the internal vapour through the solid's dry layer. Under the low temperature and with the absence of water transfer through the pores, the food matrix does not collapse and develops a significant porosity.
3. Second drying stage. This stage involves the removal of the unfrozen water by evaporation (desorption) and begins when the ice has already been removed by sublimation. The bound water is removed by heating the product under vacuum; as its removal is slower than the removal of free water it affects significantly the overall drying time. The heat supplied in this stage should be controlled because the structure of the solid matrix may undergo significant modification if the temperature of the product rises. The energy delivered to the solid can be supplied by conduction, convection and/or radiation (Voda *et al.*, 2012).

FD is a very versatile drying method but its cost is very high due to the need of freezing the raw materials and operating under high vacuum for dehydration (Claussen *et al.*, 2007; Ratti, 2001). A significant advantage of FD is the minimum change of most of the initial food material properties, such as structure, shape, appearance, texture, biological activity and nutrient compounds, and the retention of colour, flavour, aroma and taste. This is possible as the food is processed at low temperatures in the absence of air. Other advantages of FD include the ability of almost complete removal of water, the high porosity of the final product, which leads to a fast rehydration rate and high rehydration capacity, and the ability to convert the material to powder with low mechanical requirements (e.g. by adding it in an extrusion cooking feed mixture). Chemical (e.g. oxidation and modification) reactions and/or enzymatic reactions are significantly limited and vitamin degradation is reduced in comparison to classical drying techniques.

A major disadvantage of FD is the duration of the process (1 to 3 days). This is due to poor internal heat transfer inside the product and a low working pressure as the principal heat transfer phenomenon is radiation. Product characteristics, such as texture, degree of ripeness and dry matter content, and processing conditions, such as loading density, height of the product layer,

specific surface of the product and condenser capacity, are variables that have a considerable effect on the FD time, but they are also essential for the rehydration ratio and texture of the final product (Hammami and René, 1997).

FD is applicable to pharmaceuticals, biotechnology products, enzymes, nutraceuticals and other high value and quality materials. In food industry, it is restricted to high value-added products, such as coffee, tea and infusions, ingredients for ready-to-eat foods (vegetables, pasta, meat, fish, etc.) and several aromatic herbs.

Hammami and René (1997) studied the production of high-quality freeze-dried strawberry pieces. A working pressure of 30 Pa and heating plate temperature of 50 °C were the optimal conditions used to maximize the final product quality, including appearance, shape, colour, texture and rehydration ratio. Voda *et al.* (2012) investigated the impact of FD, blanching pre-treatment and freezing rate on the microstructure and rehydration properties of winter carrots by μCT (micro-computed tomography), SEM (scanning electron microscopy), MRI (magnetic resonance imaging) and NMR (nuclear magnetic resonance) techniques. It was concluded that the freezing rate determines the size of ice crystals being formed, which leave pores upon drying. The samples frozen at a lower temperature showed smaller pores as the ice crystals are expected to grow less under fast cooling conditions. During freezing, the growth of an ice crystal ruptures, pushes and compresses cells and this damage is more pronounced in slowly frozen tissue, which yields bigger ice crystals.

Duan, Ren and Zhu (2012) developed a microwave freeze drying (MFD) technique to dry apple slices. Nevertheless, MFD is a very sensitive procedure due to the inherently nonuniform distribution of the microwave field, which leads to an uneven temperature distribution in the drying material, leading to overheating and quality deterioration. Based on the dielectric properties of the material, a changed microwave loading scheme could lead to perfect product quality and greatly reduce the drying time. MFD took ~6 hours of processing time, which was ~60% less than that for conventional FD.

1.3.5 Spray drying

Spray drying (SD) is a special process used to transform a feed from a liquid state to a dried particulate form by spraying the feed into a hot drying medium. The feed can either be a solution, suspension, emulsion or paste. Different types of food materials can be produced, such as powders, granules and agglomerates at different sizes. In the SD process, the fluid is atomized using a rotating disc or a nozzle and the spray of droplets comes immediately in contact with a flow of hot drying medium, usually air. During evaporation from a small liquid droplet, moving through the turbulent body of hot fluid under the influence of gravity and its own initial kinetic energy, a complicated function of simultaneous conduction and convection of heat from the fluid to the droplet

surface, and diffusion and convection of water vapour back into the body of fluid take place. The boundary layer is separated by the interaction of the fluid with the droplet surface; its shape changes rapidly and the solute in the droplet becomes concentrated and finally solid. The rapid evaporation maintains a low droplet temperature so that a high drying air temperature can be applied without affecting the quality of the product. The drying process may last only a few seconds. The low product temperature and short drying time allow SD to process extremely heat-sensitive materials. The process is continuous and easy to be controlled, and satisfies aseptic/hygienic drying conditions. Disadvantages of the method are the relatively high cost, the low thermal efficiency and the large air volumes at low product hold-up. SD is used in the production of coffee, tea extract, tomato paste, powdered cheese eggs, enzymes (amylase used in baking and brewing, protease used in brewing, meat and fish tenderizing and cheese making, glucose oxidase used in carbonated beverages, pectinase used in coffee fermentation and juice clarification, rennin used in cheese making, lactase used in ice cream, dextranase, lipase, pepsin and trypsin), skim milk, spirulina, soups, maltodextrin, soya protein, sweeteners, etc.

A variation of SD is superheated steam spray drying, which can be used with no fire and explosion hazards, no oxidative damage, the ability to operate at vacuum or high operating pressure conditions, ease of recovery of latent heat supplied for evaporation and minimization of air pollution due to operation in a closed system. In the past few years, spray freeze drying has received much attention. It consists of the following stages:

- atomization of liquid solutions or suspensions using ultrasound, one or two fluid nozzles or vibrating orifice droplet generators,
- freezing of the droplets in a cryogenic liquid or cryogenic vapour, and
- ice sublimation at low temperature and pressure or alternatively atmospheric freeze drying using a cold desiccant gas stream.

Goula and Adamopoulos (2005) investigated the production of tomato powder by SD tomato pulp in a modified spray dryer connecting the spray dryer inlet air intake to an air dehumidifier. It was observed that the moisture content of the powder decreased with an increase in air inlet temperature and compressed air flow rate, and with a decrease in drying air flow rate. Bulk density increased with a decrease in drying air flow rate and air inlet temperature, and with an increase in the compressed air flow rate. Solubility increased with a decrease in drying and compressed air flow rate and with an increase in the air inlet temperature. Without preliminary air dehumidification, the moisture content of the powder was higher and its bulk density and solubility were lower, indicating that the rapid particulate skin formation improved the product recovery and its properties.

One of the most important applications of SD is the food encapsulation and micro-encapsulation. These techniques are an efficient way of

raising the shelf-life of food during storage. The most common materials used for micro-encapsulation using SD are gums, like gum Arabic, low-molecular-weight carbohydrates, such as maltodextrins and saccharose, cellulose, gelatine, lipids and proteins, e.g. soy proteins. Borrmanna *et al.* (2012) investigated the shelf-life of vitamin C encapsulated with *n*-octenylsuccinate (*n*-OSA)-derived starch in passion fruit juice produced by SD. SD proved itself as an inexpensive alternative to freeze drying, capable of retaining vitamin C during a long time of storage and easily diluted in cold water in order to reconstitute passion fruit juice for human consumption.

Fazaeli *et al.* (2012) studied the effects of some processing parameters on moisture content, water activity, drying yield, bulk density, solubility, glass transition temperature and microstructure of spray-dried black mulberry juice powders. The effect of SD conditions revealed that a higher inlet air temperature, increase of carrier agent concentration or decrease of maltodextrin DE caused an increase in process yield and solubility and a decrease in bulk density, moisture content and water activity. The blend of maltodextrin 6DE and gum Arabic proved to be more efficient (drying yield of 82%) than the other blends, resulting in better physical properties and powder morphology.

1.3.6 Osmotic dehydration

Osmotic dehydration (OD) is a simple and useful technique for removal of water from fruits and vegetables, realized by placing the solid food in aqueous solutions of sugars and salts possessing high osmotic pressure. The correct term to be used is 'osmotic dewatering' since the final product still has a high moisture content, a lot higher than 2.5%. During OD three simultaneous countercurrent flows occur:

- a significant amount of water flows out of the food into the solution (water loss),
- a transfer of solutes from the solution into the food (soluble solids uptake) and
- a leakage of solute molecules (hydrosolubles), such as sugars, salts, organic acids and minerals, across the membrane into the solution.

The first two flows take place due to the water and solute activity gradients across the cell's membrane, while the third one, which is minor from a quantitative point of view but may be essential as far as organoleptic or nutritional qualities are concerned, occurs due to the differential permeability of the cell membranes (Torreggiani, 1993).

Through OD, the introduction of a preservative agent or any solute of nutritional interest, which is capable of giving the product better sensory characteristics and reduced water activity, is possible (Buggnhout *et al.*, 2008). Mass transfer during OD is affected by several parameters, such as osmotic solution

composition, concentration, temperature, osmosis duration, pressure and type and extent of agitation.

OD is usually used as a pre-treatment and not for the production of dried food materials as a 30–40% reduction of food water is considered to be the optimum. Thus OD is followed by other dehydration methods like HAD, VD and FD. OD, which is effective even at ambient temperature, preserves texture and colour. The amount of water remaining in the material, however, does not ensure its stability, as water activity is generally higher than 0.9. Nevertheless, compared to fresh fruits, the osmotic-treated ones present increased microbiological stability for further processing and subsequent storage period due to sugar uptake, owing to the protective action of the saccharides. The semi-dried fruit ingredients produced by OD are included in a wide range of complex foods such as ice creams, cereals, dairy, confectionery and baking products (Tortoe, 2010). A significant advantage of OD is its low energy consumption compared to other drying methods such as HAD and FD.

Vasconcelos *et al.* (2012) studied OD of Indian fig with two binary solutions (sucrose/water and glucose/water) and a ternary solution (sucrose/NaCl/water). They found that temperature had a greater influence on the water loss, while concentration had a greater influence on the solid gain in all three hypertonic solutions investigated. The best conditions for OD of Indian fig to maximize water loss and minimize solid gain were in glucose solution of 40° Brix at 40 °C for 165 min. The properties of foods undergoing OD can be enhanced by combining osmotic treatment with other drying techniques acting simultaneously.

1.3.7 Atmospheric freeze drying

Atmospheric freeze drying (AFD) is an alternative to vacuum freeze drying (FD). The most effective method to apply AFD is by using a fluidized bed dryer. The drying rate depends on the operating temperature, pressure and material thickness. AFD is a much slower method compared to FD due to being an internally controlled mass transfer process. The drying time can be 2 to 4 times higher than FD for materials of the same dimensions, depending on the pressure in the dryer. Lower pressures and smaller dimensions of the food particles tend to reduce the drying time (Kudra and Mujumdar, 2002).

One way to apply AFD is to use a second material compatible with the food, such as starch granules or zeolite. The aim of these materials is to transfer heat for the ice sublimation and to absorb the moisture released. Both materials are in a fluidized state due to the feed of cold air. The mixture is separated and the absorbent is heated and regenerated in order to lose the excess moisture and immerse again in the dryer after cooling. Silica gel can also be used to entrap the water removed in the form of ice, following its regeneration (Reyes *et al.*, 2010).

Another practical and convenient approach is the utilization of a heat pump. Bantle, Kolsaker and Eikevik (2011) used this method to study the drying kinetics of different food materials undergoing AFD. The wet air from the drying chamber was cooled under its saturation point in the evaporator of the heat pump, which caused water to condense out. The drying air was again heated up to its working temperature in the heat pump condenser and fed back into the drying chamber. R404 was used as the refrigerant, which allowed adjustment of the drying conditions from −10 °C and relative humidity (RH) of 20–25% to 30 °C and 5% RH depending on the inlet air velocity.

Reyes *et al.* (2010) studied the drying conditions of Murtilla using AFD in a pulsed fluidized bed and vacuum FD. They concluded that in the first drying stage (sublimation) only the rate of freezing was a significant variable, which can be attributed to the generation of small ice crystals that increased the rate of drying by increasing the area of sublimation. In the second drying stage (elimination of bound water), fast freezing with infrared radiation (IR) allowed a final moisture content to be achieved that was similar to freeze-dried products in equivalent total drying periods. Slow freezing without application of IR preserved the polyphenol content better than fast freezing, whereas the antioxidant activity showed a lesser decrease with the application of IR.

Claussen *et al.* (2007) developed a simplified mathematical model (AFD-sim) based on uniformly retreating ice front (URIF) considerations to simulate industrial AFD of different foodstuffs in a tunnel dryer. The outputs from this model were the prediction of drying time, dry zone thickness and average moisture content versus relative tunnel position.

1.3.8 Sonic drying

Sonic drying depends on the energy generated in the form of sound waves. Many researchers studied the increase of drying rate in an ultrasonic field and presented a number of theories. It seems that the effect of sound in moisture removal is quite complex and caused by a decrease in viscosity, reduction of the laminar sublayer thickness due to an increase in turbulence of the air stream in contact with the material, increase of moisture evaporation due to breakage of the boundary layer and an increase in the moisture migration due to the expansion of vapour bubbles inside capillaries. Gallego Juarez (1998) concluded that diffusion at the boundary between a suspended solid and a liquid is substantially accelerated in an ultrasonic field and heat transfer is increased by approximately 30–60% depending on the intensity of the ultrasound. The mechanism of ultrasound drying (USD) is based on the principle that ultrasound travels through a medium like any sound wave, resulting in a series of compression and rarefaction. At sufficiently high power, the rarefaction exceeds the attractive forces between molecules in the liquid phase, which leads to the formation of cavitation bubbles to release energy for many chemical and mechanical effects.

Ultrasound techniques are simple, relatively cheap and energy saving, and thus became an emerging technology for probing and modifying food products. High-power (low-frequency) ultrasound modifies the food properties by inducing mechanical, physical and chemical/biochemical changes through cavitation (Kudra and Mujundar, 2002). In addition, probes that generate high power ultrasound are cheap, portable and modifiable to suit different purposes in the food industry (Awad *et al.*, 2012).

Kudra and Mujumdar (2002) presented five main sound generators applicable in the drying industry, which are: piezoelectric, magnetostrictive, electromagnetic, electrostatic and mechanical. Mechanical generators are the most common equipment used for the generation of sound in gases at frequencies up to 25 kHz, which include the Galton whistle, Hartman whistle, wedge resonator, dynamic siren, modified Hartman whistle and Branson sound generator.

The assistance of air drying with an ultrasound field can reduce drying time to about half, depending on sound energy and frequency. Sonic-assisted drying does not create hot areas inside the material and neither does it enhance moisture vaporization due to temperature increase. This is important for food drying as heat-sensitive compounds are not deteriorated by sound waves.

Nowacka *et al.* (2012) investigated the utilization of ultrasound as a mass transfer enhancing method prior to drying of apple tissue. The ultrasound treatment caused a reduction of the drying time by 31–40% in comparison to untreated tissue. Garcia-Perez *et al.* (2012) tested the feasibility of power ultrasound to intensify low-temperature drying processes for carrot, eggplant and apple cubes. The drying time was shortened by between 65 and 70%. Bantle and Eikevik (2011) used ultrasound in an AFD process of peas and concluded that the effective diffusion could be increased by up to 14.8%. The higher effective diffusion is significant for drying at low temperatures (−6 to 0 °C), whereas for higher temperatures (10 to 20 °C) the effect of ultrasound was marginally smaller.

Schössler, Thomas and Knorr (2012) studied the cellular effect of contact power ultrasound on potato cell tissue with the impact on water removal. Ultrasound-related cell disruption was limited to a thin layer (< 1 mm) directly at the sonicated surface of the potato tissue. At deeper tissue layers, structural changes were attributed to water removal.

1.3.9 Heat pump drying

Heat pump drying (HPD) is a variation of hot-air drying or generally fluid drying in which, through a mechanical arrangement, heat from the exhaust drying fluid is recovered and offered again to the moist material or even the moisture from the exhaust drying fluid is removed and the fluid recirculates in the dryer. In a heat pump dryer, which is a combination of a heat pump and a drying unit, both the latent and sensible heat can be recovered, improving the

overall thermal performance and yielding effective control of air conditions at the inlet of the dryer. Energy savings of about 40% by using heat pump dryers have been reported as compared to electrical resistance dryers (Queiroz, Gabas and Telis, 2004). Heat pump dryers used in industrial applications have been proven as drying systems that ensure the product's quality in food and agricultural products and are able to control temperature, relative humidity and velocity of the drying medium and drying duration. The heat pump has been modified to a gas engine-driven heat pump, ground source heat pump, solar heat pump, photovoltaic/thermal heat pump, chemical heat pump and desiccant heat pump (Goh *et al.*, 2011).

A few limitations of a heat pump dryer include (Daghigh *et al.*, 2010):

- the requirement of an auxiliary heating for high-temperature drying due to the critical pressure level of some refrigerants,
- the initial capital cost that may be high due to many refrigerant components,
- the requirement of a period for the system to attain desired drying conditions,
- the requirement of regular maintenance of components, and
- the leakage of refrigerant to the environment where there is a crack in a pipe due to pressurized systems.

A heat pump dryer (Figure 1.2) includes a drying cabinet, a heat pump, which consists of an evaporator, a condenser, a compressor and an expansion valve, and auxiliary equipment. Moist solids and hot air (or inert gas) are fed into the cabinet and come in contact with each other. Solids are fed on a conveyor belt of trays. Fluid circulates with a desirable velocity via an appropriate fan. Moisture is transferred from the solids to the fluid. Moist fluid passes through the heat pump evaporator and cools as the refrigerant vaporizes. The fluid becomes saturated and, as its temperature further reduces, water is removed from it and is collected as a condensate in a water collector. Fluid (gas) separates from water in this collector. Refrigerant vapours are fed to the compressor, and their pressure and temperature increase. The refrigerant is then fed into the two condensers. Through the internal condenser, the refrigerant condensates and heat is transferred to the cold fluid (gas) coming from the water collector, to be heated again and recirculated to the drying cabinet. An external condenser is also used for adjustment of the fluid temperature to the target value. The more refrigerant fed into the internal condenser, the higher is the drying fluid temperature. The external condenser usually uses cooling water for heat removal. The refrigerant from the two condensers passes through an expansion valve and its pressure reduces to the working pressure of the evaporator. An auxiliary steam heater is also shown in Figure 1.2. Under steady-state conditions, its heat duty is equal to

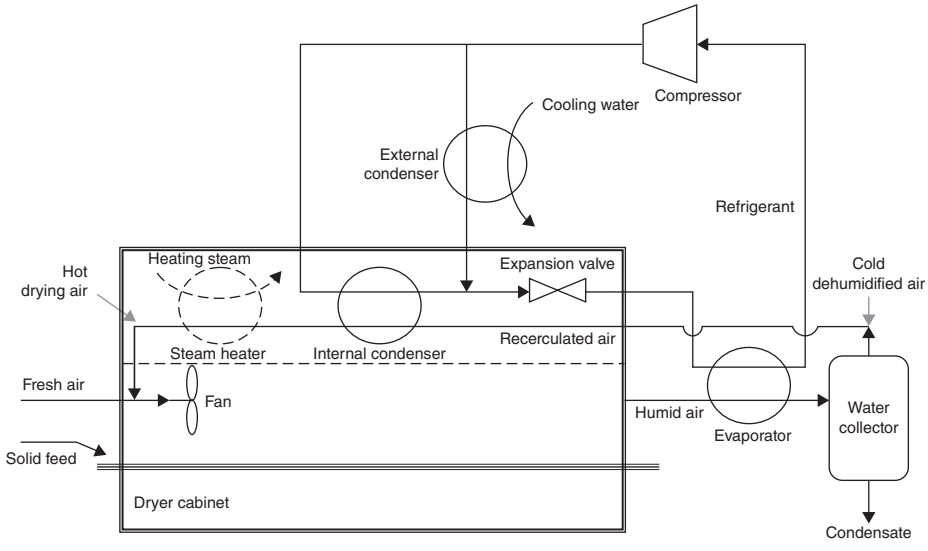

Figure 1.2 Heat pump dryer

zero. Nevertheless, its existence is necessary during the start-up of the unit as well as for its better control in case of troubleshooting of the heat pump.

Table 1.1 presents the mathematical model of the described heat pump dryer. In this model, it is assumed that the mass of dry air (or inert gas) remains constant in the dryer and under steady-state conditions the water removed from the moist solids and transferred to the drying medium inside the drying cabinet is equal to the water removed from the moist fluid in the heat pump evaporator-water collector system. Thus, the recirculated low-humidity dewatered drying medium is considered as a closed system. In a real dryer, drying fluid losses are possible and the complete recirculation may not apply. In these cases, fresh ambient air or stored inert gas also enters the dryer. The presented model can be easily modified to describe these cases, by adding the mass and energy (enthalpy) equations for a mixing point of ambient and recirculated fluid. Further, a mass balance equation should be included for the drying medium removed from the system. Table 1.2 presents the process variables of the heat pump dryer for the terms used in Table 1.1.

HPD may use inert gases like N_2 and CO_2 for food dewatering. The moisture removed from the material is collected by the inert gas used. Since the use of inert gases is much more expensive than the use of air, the inert gas should be recycled to the drying process and not rejected to the atmosphere. To be able to do this, its moisture has to be removed, as it decreases the drying rate. This can be easily achieved by cooling the fluid stream in the heat pump evaporator in order for the moisture to be liquefied and the separated inert fluid to be recirculated for further moisture removal after heating again in the

Table 1.1 Mathematical model of a heat pump dryer

$$F_w = F_s(X_i - X_o) = F_a(Y_{vo} - Y_{vi})$$

$$Y_{vo} = Y_{vi} + \frac{F_w}{F_a}$$

$$F_s(C_{ps} + X_i C_{pw})T_a + F_a(C_{pa} + Y_{vi}C_{pv})T_i = F_s(C_{ps} + X_o C_{pw})T_o + F_a(C_{pa} + Y_{vo}C_{pv})T_o + F_w(\Delta H_w + C_{pv}T_o - C_{pw}T_a)$$

$$P_{so} = \exp\left(a_1 - \frac{a_2}{a_3 + T_o}\right)$$

$$Y_{so} = \frac{mP_{so}}{P - P_{so}}$$

$$T_{di} = \frac{a_2}{a_1 - \ln\left(\frac{Y_{vi}P}{m+Y_{vi}}\right)} - a_3$$

$$Q_e = F_a\lfloor C_{pa}(T_o - T_{di}) + C_{pv}(Y_{vo}T_o - Y_{vi}T_{di}) + (Y_{vo} - Y_{vi})\Delta H_w\rfloor + F_w C_{pw}T_{di}$$

$$Q_c = F_r\left[\Delta H_r \frac{T_{ac} + 273}{T_e + 273} - C_{pr}\left(T_{ac} - T_e\right)\right]$$

$$F_r = \frac{Q_e}{\Delta H_r - C_{pr}(T_{ac} - T_e)}$$

$$E_c = F_r \Delta H_r \frac{T_{ac} - T_e}{T_e + 273}$$

$$Q_{ah} = F_a(C_{pa} + Y_{vi}C_{pv})(T_i - T_{di})$$

$$Q_r = Q_c - Q_{ah}$$

$$Y_{so} > Y_{vo}$$

heat pump condenser or/and through a different heating source (Doungporn, Poomsa-ad and Wiset, 2012). Drying under an inert atmosphere presents multiple advantages, such as:

- higher drying rate due to higher heat and mass transfer,
- absence of oxidative reactions, which is especially critical in the drying of sensitive materials present in food products (Perera and Rahman, 1997),
- reduction of browning and shrinkage, and quick rehydration (O'Neill *et al.*, 1998),
- very high overall quality, retention of vitamin C and the colour of the product similar to products obtained from vacuum or freeze drying (Hawlader, Perera and Tian, 2006), and
- decrease of temperature increments leading to superior product quality (Hawlader *et al.*, 2006).

Doungporn, Poomsa-ad and Wiset (2012) studied thin-layer drying characteristics of Thai Hom Mali paddy using different drying gases (hot air, CO_2

Table 1.2 Process variables of a heat pump dryer

a_1 (−)	Antoine constant
a_2 (−)	Antoine constant
a_3 (−)	Antoine constant
C_{pa}(kJ/kg K)	Heat capacity of dry air
C_{pr} (kJ/kg K)	Heat capacity of refrigerant
C_{ps} (kJ/kg K)	Heat capacity of solids
C_{pv}(kJ/kg K)	Heat capacity of water vapor
C_{pw}(kJ/kg K)	Heat capacity of water (liquid removed from the solids)
E_c (kW)	Refrigerant compressor power
F_a (kg (d.b.)/s)	Drying air flow rate (d.b. is dry basis)
F_r (kg/s)	Refrigerant flow rate
F_s (kg (d.b.)/s)	Dry solids flow rate (dry basis)
F_w (kg/s)	Rate of water removal from the solids
m (−)	Air/water molecular weight ratio
P (atm)	Ambient pressure, working pressure of the dryer
P_{so} (atm)	Vapour pressure at saturation of the outlet air from the dryer
Q_{ah} (kW)	Heat load added to the recirculated drying air
Q_c (kW)	Cumulative heat duty of the two heat pump condensers
Q_e (kW)	Heat duty of the heat pump evaporator
Q_r (kW)	Heat load removed from the external condenser
T_a (°C)	Ambient temperature, initial temperature of solid
T_{ac} (°C)	Heat pump condenser temperature
T_{di} (°C)	Dew point temperature of dryer cabinet inlet air, dew point temperature of air exiting heat pump evaporator
T_e (°C)	Heat pump evaporator temperature
T_i (°C)	Dryer cabinet inlet air temperature
T_o (°C)	Dryer cabinet outlet air temperature
X_i (kg/kg d.s.)	Initial moisture content of the solids (d.s. is dry solids)
X_o (kg/kg d.s.)	Final moisture content of the solids
Y_{so} (kg/kg d.a.)	Saturation absolute humidity of the dryer cabinet outlet air (d.a. is dry air)
Y_{vi} (kg /kg d.a.)	Absolute humidity of dryer cabinet inlet air
Y_{vo} (kg/kg d.a.)	Absolute humidity of dryer cabinet outlet air
ΔH_r (kJ/kg)	Latent heat of vaporization of the refrigerant
ΔH_w (kJ/kg)	Latent heat of vaporization of water (at 0 °C, reference temperature)

and N_2) at a temperature range 40–70 °C in a heat pump dryer. The drying rate was not affected by the drying medium but increased with the drying temperature. The Midilli model in the form of the Arrhenius type was the best model for describing the drying behaviour of the product. Figure 1.3 presents the flowsheet of the heat pump and Table 1.3 summarizes the flow, composition and properties of each stream. The phase equilibrium has been calculated through thermodynamic models. One of the most interesting results of this simulation is the dilution of a small amount of nitrogen to the cold water removed from the water collector. An addition of ~5.7 kg per day of N_2 is

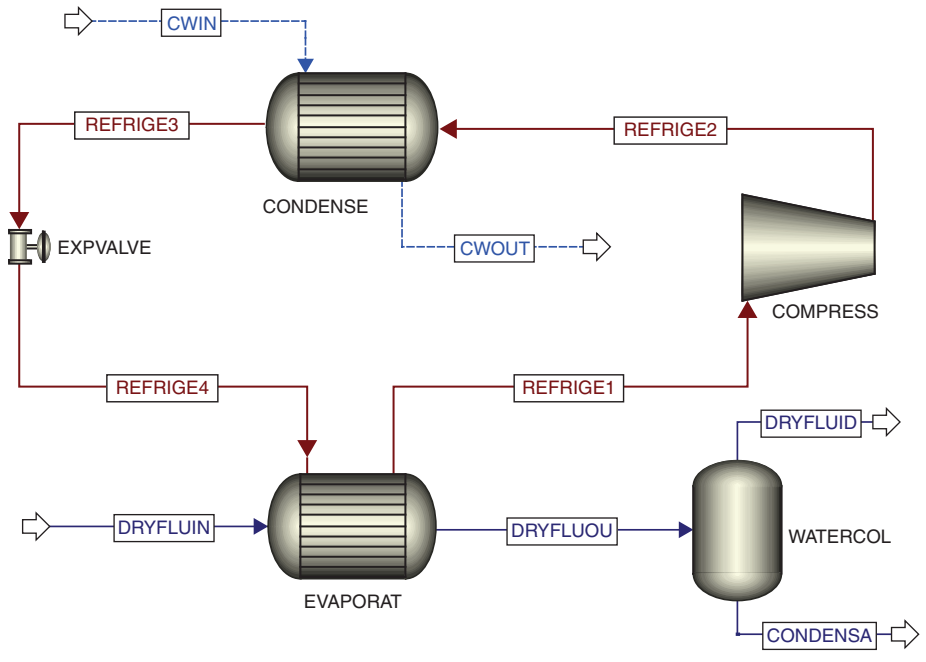

Figure 1.3 Heat pump for the dewatering of nitrogen–water vapour mixture

necessary to cover the losses. Furthermore, the water separated in the water collector is cold and has to be heated fast before freezing. Table 1.4 presents some useful performance data.

1.3.10 Infrared drying

Infrared drying (IRD) is an emerging method presenting significant advantages, including a relatively shortened drying time, high energy transfer rate and therefore high drying efficiency, reduced energy consumption, efficient transmission through the air or evacuated space, lower air flow through the product, uniform temperature in the product, superior product quality, space saving, ease of automation and a clean working environment compared to other drying methods. IRD is based on the interaction of infrared wavelength radiation from a source with the internal structure of a food material. IR radiation impinges on the moist material to be dried, penetrates it and the radiation energy is converted into heat. The increased temperature in the inner layers of the material results in an increase of vapour pressure, which promotes moisture migration to its surface and the removal by the surrounding ventilating air (Khir *et al.*, 2012). IR energy is transferred from a heating element to the product without heating the surrounding air, resulting in a higher temperature in the inner layers of the product compared to the air. During the first period of

Table 1.3 Properties of the streams of the heat pump in Figure 1.3

Stream	Drying fluid before cooling	Drying fluid after cooling	Dry fluid	Condensate	Refrigerant exiting evaporator	Refrigerant exiting compressor	Refrigerant exiting condenser	Refrigerant exiting expansion valve	Cooling water inlet	Cooling water outlet
Stream symbol	DRYFLUIN	DRYFLUOU	DRYFLUID	CONDENSA	REFRIGE1	REFRIGE2	REFRIGE3	REFRIGE4	CWIN	CWOUT
Temperature (°C)	65	−13.2	−13.2	−13.2	−17.5	48	37.6	−17.5	20	31.4
Pressure (bar)	1.013	1.013	1.013	1.013	0.507	3.546	3.546	0.507	1.013	1.013
Vapour fraction (−)	1	0.956	1	0	1	1	0	0.354	0	0
Mole flow (kmol/h)	130.6	130.6	124.9	5.73	37.2	37.2	37.2	37.2	999.1	999.1
Mass flow (kg/h)	3600	3600	3496.6	103.3	2160	2160	2160	2160	18000	18000
Volume flow (m^3/h)	3625.1	2664.3	2664.2	0.1	1559.2	279.7	3.87	554.24	18.022	18.223
Enthalpy (MMkcal/h)	−0.31	−0.443	−0.049	−0.394	−1.152	−1.096	−1.285	−1.285	−68.259	−68.07
	Mass flow (kg/h)									
N-BUT-01	0	0	0	0	2160	2160	2160	2160	0	0
WATER	108	108	4.9	103.08	0	0	0	0	18 000	18000
NITRO-01	3492	3492	3491.7	0.237	0	0	0	0	0	0

Mass frac (–)										
N-BUT-01	0	0	0	0	1	1	1	1	0	0
WATER	0.03	0.03	0.001	0.998	0	0	0	0	1	1
NITRO-01	0.97	0.97	0.999	0.002	0	0	0	0	0	0
Mole flow (kmol/h)										
N-BUT-01	0	0	0	0	37.16	37.16	37.16	37.16	0	0
WATER	5.99	5.99	0.273	5.722	0	0	0	0	999.1	999.1
NITRO-01	124.6	124.6	124.6	0.008	0	0	0	0	0	0
Vapour phase										
Enthalpy (cal/gm)	−86.003	−14.009	−14.009		−533.112	−507.33		−533.113		
Heat capacity (cal/gm K)	0.255	0.248	0.248		0.36	0.43		0.36		
Conductivity (kcal m/h m^2)	0.024	0.02	0.02		0.01	0.016		0.01		
Liquid phase										
Enthalpy (cal/g)		−3813.1		−3813.1			−594.8	−628.7	−3792.1	−3781.6
Heat capacity (cal/gm K)		0.883		0.883			0.678	0.56	0.913	0.931
Conductivity (kcal m/h m^2)		0.422		0.422			0.088	0.107	0.515	0.529

Table 1.4 Performance data of the equipment

Condenser	
Heat duty (Gcal/h)	0.18902
Logarithmic mean temperature difference (°C)	11.36
Evaporator	
Heat duty (Gcal/h)	0.133353
Logarithmic mean temperature difference (°C)	21.91
Compressor	
Isentropic efficiency (–)	0.72
Mechanical efficiency (–)	0.75
Indicated horsepower (kW)	64.8
Net work required (kW)	86.4
Isentropic power requirement (kW)	46.6
Power loss (kW)	21.6
Calculated pressure ratio (–)	7
Outlet temperature (°C)	48
Isentropic outlet temperature (°C)	37.6
Head developed (m)	7925
Inlet heat capacity ratio (–)	1.105

drying when the sample surface is coated with a very thin layer of water, the IR is extraordinarily energy efficient and speeds up the drying process (Kowalski and Mierzwa, 2011). The drying takes place from inner to outer layers via both radiation and convection phenomena. IRD is particularly valid for products with a significant moisture content, for which long-wave radiation (over 3 μm) is almost totally absorbed by moisture as there is a very good correlation of IR wavelengths with the absorption bands of water, while dry material is highly permeable to such radiation.

Niamnuy *et al.* (2012) studied the drying of soybean, and the interconversion and degradation of soy isoflavones during gas-fired infrared combined with hot-air vibrating drying (GFIR–HAVD). The de-esterification is the predominant reaction of isoflavone changes during drying, and the conjugated glucosides have less stability than the aglycones form of isoflavones. GFIR–HAVD at 150 °C gave the highest drying rate and conversion rates of various glucosides to aglycones. However, the high degradation rates of all isoflavones occurred at a temperature of 150 °C and hence a drying temperature of 130 °C is recommended as the most suitable temperature to optimize drying rate, conversion rates of various glucosides to aglycones and degradation rates of all isoflavones. Kowalski and Mierzwa (2011) studied the hybrid drying of microwave, infrared and hot-air drying and came to the conclusion that MWD is enhanced for red bell pepper; the process consisted of eight phases. In phases 1, 3, 5 and 7, the process was enhanced with IR.

A phase was terminated when the temperature attained a specified level. In phases 2, 4 and 6, only the microwave (MW) energy was supplied. Shrinkage and deformation of samples were smaller, the aroma was better preserved and colour was conserved to a satisfactory degree compared to hot-air-dried products, while energy consumption was also smaller.

1.3.11 Superheated steam drying

Superheated steam drying (SSD) is an emerging technology, which uses superheated steam as the drying medium for heat supply to a product to be dried and carry off evaporated moisture instead of hot air as used in HAD. This equipment is more complex than a hot-air dryer. There is no fire or explosion hazard. The application of low, near-atmospheric (preferable due to reduced equipment cost) or high pressure (at ~5 bar, referred to as high pressure superheated steam drying) operation is possible. Any convection dryer such as fluidized bed, flash, rotary, conveyor type, etc., can be transformed into superheated steam dryer, while additional heat sources (radiation, microwave, etc.) can also be combined. The net energy consumption can be low enough (up to 80% reduction) when integration systems are applied for the heat recovery (Raghavan *et al.*, 2005). Thermal properties of steam are superior compared to air at the same temperature, resulting in a higher heat transfer coefficient. Furthermore, vapour transfer is faster than liquid diffusion, thus improving mass transfer during drying as well. Since the drying medium does not contain oxygen, there is no risk of oxidation of food substances (enzymatic browning and lipid oxidation) and the product quality is quite good, including the preservation of nutrients and colour, although heat-sensitive materials may be prone to damage. Case-hardened skin is unlikely to be formed in this method and the treatment strips out more of the acids that contribute to an undesirable taste or aroma of the products. Pasteurization, sterilization, deodorization or other heat treatments (e.g. blanching, boiling, cooking) of the product may take place simultaneously with drying and the product presents higher porosity due to evolution of steam within the product as boiling in the interior opens up the elastic wet solid. This results in lower bulk density and better rehydration behaviour.

SSD has been applied successfully to many food materials, including potato chips, tortilla chips, shrimp, paddy, soybean, Asian noodles, pork, chicken, fermented fish, sugar beet pulp, spent grain from a brewery, okara, sunflower seed, cacao bean and pressed beet pulp after extraction of sugar, where high pressure superheated steam drying can de used.

A variant of SSD is low-pressure superheated steam drying (LPSSD), which is used in the cases where products to be dried melt, undergo glass transition or are damaged at the saturation temperature of steam. This

method takes place at a pressure of 5–10 kPa, combines the ability to dry the product at a low temperature with some advantages of SSD and leads in product quality preservation and an enhanced drying rate. It is suitable for highly heat-sensitive products such as herbs, fruits, vegetables, edible films, functional foods and ingredients, as well as other bioactive materials for which the SSD is prohibited.

Devahastin *et al.* (2004) used carrot cubes as a model heat-sensitive material to investigate various quality parameters of the dried product undergoing LPSSD or VD. They recorded that convective heat transfer was poorer under reduced pressures, leading to lower drying rates, but the quality of the dried product was superior compared to that obtained using conventional VD. The effect of the operating pressure was less significant than that of the steam temperature. The shrinkage patterns resulting from LPSSD and VD processes were quite different even though the values of shrinkage were similar. Steam drying provided a much better rehydration capability of the food due to the formation of less dense layers compared to VD.

LPSSD can be combined with other dehydration methods to improve product quality. Nimmol *et al.* (2007) studied the effect of LPSSD and far-infrared radiation (FIR) on drying of banana slices. The LPSSD–FIR dried banana showed more crispness than the VD–FIR banana, especially at higher drying temperatures. LPSSD–FIR at 90 °C required a shorter drying time than VD–FIR but the colour was darker. Furthermore, the dried banana slices had higher values of colour changes compared with those that underwent LPSSD alone.

1.3.12 Intermittent drying

Intermittent drying (ID) is a drying technique in which the heat is applied in a noncontinuous way. In the tempering period, moisture is redistributed inside the material. This produces two effects:

- a quality increase (avoiding cracking of material), and
- an increase in the drying rate when heat application is rebooted (Holowaty, Ramallo and Schmalko, 2012).

A reduction in energy consumption is expected in the industrial drying of food materials when this technique is applied.

ID has been successfully applied in the dewatering of many products such as rice, banana, guava, potato, soybean and wheat. Holowaty, Ramallo and Schmalko (2012) investigated the dehydration of yerba maté branches in a bed dryer using two different tempering periods of 15 and 30 min, and concluded that both periods produced the same effect and the final moisture content of the product was practically similar to that of continuous drying. Estürk (2012) studied the drying of sage herb taking into consideration the thermal damage

during drying. He used MWD–air drying (AD) (continuous and intermittent) and convective HAD of sage to determine their effect on colour and essential oil content. The continuous MWD–AD had the fastest drying rate. The drying time of the HAD was about 63.5 to 82.4 times longer than that of the continuous MWD–AD and about 17.0 to 31.6 times longer compared to the intermittent MWD–AD depending on the mode used.

1.3.13 Instant controlled pressure drop drying

Instant controlled pressure drop (DIC, from the French *détente instantanée contrôlée*) is a drying technology from the 1980s as a treatment using a high temperature (up to 180 °C) and short time (usually less than 60 s) followed by an instant pressure drop towards a vacuum (with a pressure drop rate > 0.5 MPa/s and a pressure of approximately 5 kPa), which allows the water to abruptly autovaporize, causing controlled expansion of the product. Usually a first stage of partial drying takes place, decreasing the product moisture content to 0.2–0.3 kg H_2O/kg dry matter (d.m.), before submitting it to DIC treatment, followed by HAD for 1–2 hours. The DIC treatment usually starts by creating a vacuum condition, followed by injecting steam to the material, which keeps in contact for several seconds and then proceeds to apply a sudden pressure drop towards a vacuum. The application of DIC with HAD in fruits and vegetables can lead to lightly or highly expanded products. DIC treatment has to be applied to low moisture content products in order to act as closely as possible to the glass transition zone, which allows the expansion to be maintained. Products undergoing DIC can be of a snack type and can easily be crushed, leading to expanded granule powders with quality attributes higher than traditionally dried or spray-dried powders. This method is used for texturing fruits, vegetables and seaweeds, and presents many advantages such as reduction of energy consumption and overall production cost, controllability, improvement of the quality in terms of sensorial, functional, convenience and nutritional attributes, increased safety and hygiene, as it causes perfect decontamination due to its thermal (high temperature) and micromechanical (instant pressure drop) effects, as well as enhancement of mass transfer as it creates an open cell structure. The purpose of the texturing step is to modify the texture of food material, to improve its quality, including physical properties, and to intensify functional behaviour. Texturing comes in the form of material swelling, which leads to increased porosity and specific surface area and reduced diffusion resistance of moisture during the final dehydration step (Mounir and Allaf, 2008).

1.3.14 Sun drying and solar drying

Sun drying is the most ancient drying method, using the sun radiation to remove water from a product. It applies mainly in agricultural and farming

food materials in places where the insolation is high and the outdoor temperature is higher than 30 °C. It is a simple, costless method but drying time is very high (up to 10 days) and there is a need for extensive land. In general, the product quality is poor due to enzymatic and Maillard reactions, pigment degradation, caramelization and ascorbic acid oxidation (Kowalski and Mierzwa, 2011). Weather conditions often preclude the use of sun drying because of spoilage due to rehydration during unexpected rains. Direct exposure to the sun might cause case hardening, while marauding animals and insects, contamination by pests and the growth of toxic fungi can cause loss of a product portion.

Solar drying reclaims the sun's energy by controlling the radiative heat. Solar drying decreases drying time, increases efficiency due to reduced harvest losses, retains more of the nutritional value as drying takes place at an optimum temperature and produces significant cost savings by reducing conventional fuel demand. It has been applied in grains, fruits, meat, vegetables, fish, etc. Solar drying can be achieved by direct exposure to the sun's radiation or by incorporating external means, such as fans, to transfer solar energy in the form of heated air from the collector area to the drying chambers. Products dried by the first technique include banana, pineapple, mango, carrots, etc., while the latter is used for drying higher moisture content foodstuffs such as papaya, kiwi fruits, cabbage and others. A coal stove or agricultural wastes can be incorporated as auxiliary heating sources, while sometimes wood smoke can be used for drying (Chua and Chou, 2003).

1.3.15 Supercritical drying

Supercritical drying (SCD) is an emerging drying method which results not simply in water removal from a food material but, furthermore, in the retention of its original (micro) structure and the corresponding porous network functionality (Brown *et al.*, 2010, 2008). When SCD is applied at appropriate operating conditions, a single homogeneous phase can be formed between the supercritical fluid and the co-solvent. If used, there are no vapour–liquid interfaces and the food to be dried does not suffer from surface tension forces and capillary-induced tensile stresses, which generally damage and cause the collapse of the structure. CO_2, which is the most commonly used supercritical fluid, has a low critical temperature (~31 °C) and the drying can be realized in a near to ambient temperature. Supercritical carbon dioxide ($scCO_2$) is a non-polar solvent and the solubility of water in it is very low. Ethanol, ethyl acetate and other organic solvents when added in small quantities to $scCO_2$ can significantly increase the solubility of polar substrates in $scCO_2$. The organic solvent causes a displacement of water and the solvent/water mixture is removed with $scCO_2$. The equipment for this process is more complicated compared to conventional drying techniques.

Brown *et al.* (2008) investigated supercritical carbon dioxide drying ($scCO_2D$) of carrot by applying X-ray microtomography and light microscopy. The experiments were carried out at 20 MPa pressure using ethanol as the co-solvent. Carrots dried in the $scCO_2$–EtOH environment retained their shape much better and presented less dense structures compared to air-dried carrots, which underwent shrinkage. Brown *et al.* (2010) also examined the SCD for water removal from agar gels and compared this method to HAD and FD. They observed that for formulations containing sucrose, which displayed the best structural retention, voidage was found to increase in the order: HAD (4% voidage) < $scCO_2$ (48%) < $scCO_2$–EtOH (68%) < FD (76%).

1.3.16 Flash drying

Drying of fruits and vegetables can be realized with the application of another drying technique, which is the convective multiflash drying (CMFD) process. This process is based on the application of successive cycles of heating and vacuum pulses. The product is heated at atmospheric pressure using hot air, which causes partial dehydration of the product. When the product reaches the desired temperature, a sudden pressure reduction is applied, which leads to water evaporation (flash drying) and product cooling. Additional heating–vacuum pulse cycles can be applied to achieve the desired characteristics of the dried product. During water removal, the product undergoes texturization. As it is possible to apply many cycles, the heating temperature can be compatible with the food's sensitivity to heat treatment. This method allows the production of dehydrated fruits with moisture content, water activity and mechanical properties similar to those observed in commercial freeze-dried fruits. CMFD is an efficient dehydration technique that can be completed in shorter times (3–4 hours) and at lower capital costs, with simpler equipment and less energy requirements than FD. Banana and mango processed by CMFD were at least as crispy as the freeze-dried fruits and the colour was well preserved due to the use of moderate temperatures (Zotarelli, Porciuncula and Laurindo, 2012).

1.3.17 Pulse drying

High-temperature–short-time (HTST) pulse drying is a method used to alter the structural properties of the dried material by changing the values of the drying parameters used in conventional HAD. Hofsetz *et al.* (2007) studied the effect of the HTST pulse on HAD of banana slices and compared the properties of the material with those obtained with the HAD process. The different drying treatments led to distinctive structural changes in the food, affecting its porosity and shrinkage. The combined HTST–HAD process simultaneously puffed and dried the banana slices, resulting in reduced shrinkage compared to

the air-dried samples. For air-dried samples, the increase in porosity reached a value of 32% while during the HTST–HAD process the porosity increment reached values of 45–53% at the end of drying, resulting in the formation of a highly porous structure, which occurred together with an expansion in volume. The HTST pulse for the puffing of banana ranged from 130 to 150 °C for 23–12 min, respectively, succeeded by HAD at 70 °C. It was observed that the dehydrated bananas produced by HTST–HAD presented a crust on the external surface and big pores inside the samples, and it is believed that this crust offered resistance to shrinkage. The air-dried samples showed no crust formation and a medium–small pore structure.

The HTST drying pulse combined with HAD represents an alternative to eliminate the preceding blanching stage and promote better sensory characteristics to the final product, especially those associated with crispness. Following HTST pulse drying, the food must be further air dried to reduce the water activity to a value that will inhibit the growth of pathogenic and spoilage microorganisms, and reduce enzyme activity and the rate at which undesirable chemical and deterioration reactions occur.

1.3.18 Pulse combustion drying

Pulse combustion drying (PCD) is an emerging method based on intermittent (pulse) combustion of a solid, liquid or gaseous fuel. Such periodic combustion generates intensive pressure, velocity and, to a certain extent, temperature waves propagated from the combustion chamber via a diffuser to the dryer. Pulse combustion intensifies the rates of heat and mass transfer due to the oscillatory nature of the momentum transfer. The increased drying rates result from the impact of the sound pressure waves, which separate the surface moisture from the material undergoing drying by breaking the cohesion between the water molecules and solid particles. This greatly increases the surface area of the particles and causes rapid evaporation of water. The residence time of dispersed material is less than 5 milliseconds, which allows the drying of even thermally labile products. Pulse drying with special modified equipment has been applied in vitamins, yeast, spices, vegetable protein, fibres, whole eggs, food colourings, caramel and biopesticides, presenting quality and unit cost comparable to spray drying (Kudra and Mujumdar, 2002).

1.4 Conclusions

Drying is one of the most ancient and important processes for preservation, processing and distribution of foods and biological materials. Freeze drying is the most versatile drying method concerning the final product quality but its application is limited due to high operating costs. Hot-air drying, one of the oldest drying methods, still remains the workhorse of the drying technology.

Traditional methods, like vacuum drying, as well as emerging technologies, such as infrared drying, sonic drying and atmospheric freeze drying, aim to improve the quality compared to hot-air drying by keeping the cost at low levels. On the other hand, there are methods such as osmotic drying and instant controlled pressure drop (DIC) drying that target primarily in the compositional change or texturization of the product as they reduce the moisture content. The trend of food drying, as becomes evident from recent studies, is the development of hybrid methods, which combine advantages of two or more individual techniques to achieve the best possible quality and the most efficient energy utilization.

Abbreviations

AFD	Atmospheric freeze drying
DIC	Instant controlled pressure drop (*détente instantanée contrôlée* in French)
FD	Freeze drying
HAD	Hot-air drying
HPD	Heat pump drying
ID	Intermittent drying
IRD	Infrared drying
MWD	Microwave drying
OD	Osmotic dehydration
PCD	Pulse combustion drying
SD	Sonic drying
SSD	Superheated steam drying
VD	Vacuum drying

References

Awad, T.S., Moharram, H.A., Shaltout, O.E., Asker, D. and Youssef, M.M. (2012) Applications of ultrasound in analysis, processing and quality control of food: a review. *Food Research International*, **48**, 410–427.

Bantle, M. and Eikevik, T.M. (2011) Parametric study of high-intensity ultrasound in the atmospheric freeze drying of peas. *Drying Technology*, **29**, 1230–1239.

Bantle, M., Kolsaker, K. and Eikevik, T.M. (2011) Modification of the Weibull distribution for modeling atmospheric freeze-drying of food. *Drying Technology*, **29**, 1161–1169.

Borrmanna, D., Pierucci, A.P.T.R., Leite, S.G.F. and Leãoa, M.H.M.d.R. (2012) Microencapsulation of passion fruit (*Passiflora*) juice with *n*-octenylsuccinate-derivatised starch using spray-drying. *Food and Bioproducts Processing*, **91**(1), 23–27.

Brown, Z.K., Fryer, P.J., Norton, I.T., Bakalis, S. and Bridson, R.H. (2008) Drying of foods using supercritical carbon dioxide – Investigations with carrot. *Innovative Food Science and Emerging Technologies*, **9**, 280–289.

Brown, Z.K., Fryer, P.J., Norton, I.T. and Bridson, R.H. (2010) Drying of agar gels using supercritical carbon dioxide. *Journal of Supercritical Fluids*, **54**, 89–95.

Buggnhout, S.V., Grauwet, T., Loey, A.V. and Hendrickx, M. (2008) Use of pectin-methylesterase and calcium in osmotic dehydration and osmodehydrofreezing of strawberries. *European Food Research Technology*, **226**, 1145–1154.

Chua, K.J. and Chou, S.K. (2003) Low-cost drying methods for developing countries. *Trends in Food Science and Technology*, **14**, 519–528.

Claussen, I.C., Andresen, T., Eikevik, T.M. and Strømmen, I. (2007) Atmospheric freeze drying – Modeling and simulation of a tunnel dryer. *Drying Technology*, **25**, 1959–1965.

Daghigh, R., Ruslan, M.H., Sulaiman, M.Y. and Sopian, K. (2010) Review of solar assisted heat pump drying systems for agricultural and marine products. *Renewable and Sustainable Energy Reviews*, **14**, 2564–2579.

Devahastin, S., Suvarnakuta, P., Soponronnarit, S. and Mujumdar, A.S. (2004) A comparative study of low-pressure superheated steam and vacuum drying of a heat-sensitive material. *Drying Technology*, **22**, 1845–1867.

Doungporn, S., Poomsa-ad, N. and Wiset, L. (2012) Drying equations of Thai Hom Mali paddy by using hot air, carbon dioxide and nitrogen gases as drying media. *Food and Bioproducts Processing*, **90**, 187–198.

Duan, X., Ren, G.Y. and Zhu, W.X. (2012) Microwave freeze drying of apple slices based on the dielectric properties. *Drying Technology*, **30**, 535–541.

Dueik, V., Marzullo, C. and Bouchon, P. (2013) Effect of vacuum inclusion on the quality and the sensory attributes of carrot snacks. *LWT – Food Science and Technology*, **50**, 361–365.

Estürk, O. (2012) Intermittent and continuous microwave-convective air-drying characteristics of sage (*Salvia officinalis*) leaves. *Food Bioprocess and Technology*, **5**, 1664–1673.

Fazaeli, M., Emam-Djomeh, Z., Kalbasi Ashtari, A. and Omid, M. (2012) Effect of spray drying conditions and feed composition on the physical properties of black mulberry juice powder. *Food and Bioproducts Processing*, **90**, 667–675.

Gallego Juarez, J.A. (1998) Some applications of air-borne power ultrasound to food processing. In: Povey, M.J.W. and Mason, T.J. (eds), *Ultrasound in Food Processing*, Blackie, Glasgow, pp. 127–143.

Garcia-Perez, J.V., Carcel, J.A., Riera, E., Rosselló, C. and Mulet, A. (2012) Intensification of low-temperature drying by using ultrasound. *Drying Technology*, **30**, 1199–1208.

Goh, L.J., Othman, M.Y., Mat, S., Ruslan, H. and Sopian, K. (2011) Review of heat pump systems for drying application. *Renewable and Sustainable Energy Reviews*, **15**, 4788–4796.

Goula, A.M. and Adamopoulos, K.G. (2005) Spray drying of tomato pulp in dehumidified air: II. *The effect on powder properties. Journal of Food Engineering*, **66**, 35–42.

Hammami, C. and René, F. (1997) Determination of freeze-drying process variables for strawberries. *Journal of Food Engineering*, **7**, 133–154.

Hawlader, M.N.A., Perera, C.O. and Tian, M. (2006) Properties of modified atmosphere heat pump dried foods. *Journal of Food Engineering*, **74**, 387–402.

Hawlader, M.N.A., Perera, C.O., Tian, M. and Yeo, K.L. (2006) Drying of guava and papaya: impact of different drying methods. *Drying Technology*, **24**, 77–87.

Hofsetz, K., Lopes, C.C., Hubinger, M.D., Mayor, L. and Sereno, A.M. (2007) Changes in the physical properties of bananas on applying HTST pulse during air-drying. *Journal of Food Engineering*, **83**, 531–540.

Holowaty, S.A., Ramallo, L.A. and Schmalko, M.E. (2012) Intermittent drying simulation in a deep bed dryer of yerba maté. *Journal of Food Engineering*, **111**, 110–114.

Hottot, A., Vessot, S. and Andrieu, J. (2004) A direct characterization method of the ice morphology. Relationship between mean crystals size and primary drying times of freeze-drying processes. *Drying Technology*, **22**, 2009–2021.

Khir, R., Pan, Z., Thompson, J.F., El-Sayed, A.S., Hartsough, B.R. and El-Amir, M.S. (2012) Moisture removal characteristics of thin layer rough rice under sequenced infrared radiation heating and cooling. *Journal of Food Processing and Preservation*, **38**(1), 430–440.

Kowalski, S.J. and Mierzwa, D. (2011) Hybrid drying of red bell pepper: energy and quality issues. *Drying Technology*, **29**, 1195–1203.

Kudra, T. and Mujumdar, A.S. (2002) *Advanced Drying Technologies*, Marcel Dekker, Inc. New York.

Lee, J.H. and Kim, H.J. (2009) Vacuum drying kinetics of Asian white radish (*Raphanus sativus* L.) slices. *LWT – Food Science and Technology*, **42**, 180–186.

Lewicki, P.P. (2006) Design of hot air drying for better foods. *Trends in Food Science and Technology*, **17**, 153–163.

Mounir, S. and Allaf, K. (2008) Three-stage spray drying: new process involving instant controlled pressure drop. *Drying Technology*, **26**, 452–463.

Niamnuy, C., Nachaisin, M., Poomsa-ad, N. and Devahastin, S. (2012) Kinetic modelling of drying and conversion/degradation of isoflavones during infrared drying of soybean. *Food Chemistry*, **133**, 946–952.

Nimmol, C., Devahastin, S., Swasdisevi, T. and Soponronnarit, S. (2007) Drying of banana slices using combined low-pressure superheated steam and far-infrared radiation. *Journal of Food Engineering*, **81**, 624–633.

Nowacka, M., Wiktor, A., Śledź, M., Jurek, N. and Witrowa-Rajchert, D. (2012) Drying of ultrasound pretreated apple and its selected physical properties. *Journal of Food Engineering*, **113**, 427–433.

O'Neill, M.B., Rahman, M.S., Perera, C.O., Smith, B. and Melton, L.D. (1998) Colour and density of apple cubes dried in air and modified atmosphere. *International Journal of Food Properties*, **1**, 197–205.

Perera, C.O. and Rahman, M.S. (1997) Heat pump drying. *Trends in Food Science and Technology*, **8**(3), 75.

Queiroz, R., Gabas, A.L. and Telis, V.R.N. (2004) Drying kinetics of tomato by using electric resistance and heat pump dryers. *Drying Technology*, **22**, 1603–1620.

Raghavan, G.S.V., Rennie, T.J., Sunjka, P.S., Orsat, V., Phaphuangwittayakul, W. and Terdtoon, P. (2005) Overview of new techniques for drying biological materials with emphasis on energy aspects. *Brazilian Journal of Chemical Engineering*, **22**, 195–201.

Ratti, C. (2001) Hot air and freeze-drying of high-value foods: a review. *Journal of Food Engineering*, **49**, 311–319.

Reyes, A., Bubnovich, V., Bustos, R., Vásquez, M., Vega, R. and Scheuermann, E. (2010) Comparative study of different process conditions of freeze drying of 'Murtilla' berry. *Drying Technology*, **28**, 1416–1425.

Schössler, K., Thomas, T. and Knorr, D. (2012) Modification of cell structure and mass transfer in potato tissue by contact ultrasound. *Food Research International*, **49**, 425–431.

Šumić, Z., Tepić, A., Vidović, S., Jokić, S. and Malbaša, R. (2013) Optimization of frozen sour cherries vacuum drying process. *Food Chemistry*, **136**, 55–63.

Torreggiani, D. (1993) Osmotic dehydration in fruit and vegetable processing. *Food Research International*, **26**, 59–68.

Tortoe, C. (2010) A review of osmodehydration for food industry. *African Journal of Food Science*, **4**(6), 303–324.

Vadivambal, R. and Jayas, D.S. (2007) Changes in quality of microwave-treated agricultural products – A review. *Biosystems Engineering*, **98**, 1–16.

Vasconcelos, J.L.L.A., Andrade, S.A.C., Maciel, M.I.S., Guerra, N.B. and Vasconcelos, M.A.S. (2012) Osmotic dehydration of the Indian fig (*Opuntia ficus indica*) with binary and ternary solutions. *International Journal of Food Science and Technology*, **47**, 2359–2365.

Voda, A., Homan, N., Witek, M., Duijster, A., van Dalen, G., van der Sman, R., Nijsse, J., van Vliet, L., van As, H. and van Duynhoven, J. (2012) The impact of freeze-drying on microstructure and rehydration properties of carrot. *Food Research International*, **49**(2), 687–693.

Zotarelli, M.F., Porciuncula, B.D.A. and Laurindo, J.B. (2012) A convective multi-flash drying process for producing dehydrated crispy fruits. *Journal of Food Engineering*, **108**, 523–531.

2

Size Reduction Practices in Food Processing

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2.1 Introduction

Raw materials often occur in different sizes, varying from too large to too small to be used in the food industries. Large size samples must be reduced to an appropriate size by application of force to meet the end use requirements. In this unit operation, food is shaped into large or small pieces of certain or random shapes, or it is reduced to particles down to the micrometre range. The size reduction operation is classified into two major categories depending on the state of the material and whether a solid or a liquid. The size reduction is called grinding and cutting if it is solid. During emulsification and atomization, the size of liquid particle globules is reduced. Reaction to shear forces within solids and liquids decides the type of size reduction. Size reduction may help in fulfilling objectives like accelerating heat and mass transfer, facilitating separation of different parts of a material and obtaining pieces and particles of defined sizes for convenience to consumer and industries.

2.1.1 Size reduction of solids

Compression and shear are the two types of force commonly used in size reduction of solids, while impact forces are also used sometimes as a third type of force. Basically, the impact force is a strong compression force applied for a very short time. These forces can induce elastic and plastic deformations, and

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depend on the type of material. Beyond a certain limit, deformation results in breaking particles along certain planes. Usually, all these forces are used to different extents during size reduction, employing several methods of size reduction. They can be classified based on the final particle size, the kind of forces applied and the extent of the forces applied.

The classification, based on the final size is:

1. Breaking: > 0.15 cm
2. Cutting: 0.15 cm–8 mm
3. Crushing: 8 mm–750 μm
4. Grinding/milling: 750 μm–50 μm
5. Fine milling: < 50 μm.

Based on the force applied, the classification is:

1. Compression (e.g. pressing)
2. Shear (e.g. attrition mill, cutting)
3. Impact (e.g. hammer mill)
4. Bending (combination of compression and tension)
5. Punching (combination of compression and shear, e.g. gyratory crusher).

Based on the application of forces, the classification is:

1. Pressure and/or friction
2. Shear force on material
3. Collision between particles or impact between particles and tools
4. Friction through medium surrounding the particles.

Size reduction is important in food processing due to (a) an increased surface area, which facilitates heat exchange, chemical and biological reactions and extraction (e.g. seed oil extraction, blanching, sterilization, freezing and acidification), and (b) facilitating mixing and blending (e.g. confectionery, spices, etc.). However, some of the negative features of size reduction process are (a) energy intensive unit operation (the finer the final particle size, the higher the expenditure), (b) cutting precision significantly increases the cost of equipment, (c) heat generated in the grinding zone results in bleaching/oxidation of flour during storage, (d) loss of nutrients due to temperature rise in grinding excepting cryogenic grinding (e.g. heat-sensitive vitamins), (e) uneven particle size distribution and (f) the process is often inefficient.

Grinding Mechanical action reduces the size of the solid materials during grinding by dividing them into smaller particles. Perhaps the most extensive application of grinding is milling of grains to make flour in food industries. Also, it is widely used in many other processes, such as grinding of corn in

corn milling industries, grinding of sugar in sugar processing and confectionery plants, and size reduction of dehydrated vegetables for industries processing vegetables.

Cutting Cutting breaks down large pieces of raw materials and food into smaller pieces by mechanical action for further processing in vegetable, meat, fish and fruit processing industries. For example, a large piece of vegetable like cabbage or potato is cut to a size that fits into the can for the manufacture of canned cabbage/potato or retortable pouches containing cut pieces of different fruits and vegetables.

2.1.2 Process of grinding

In the grinding process, fracturing of materials reduces size. Even though grinding processes are widely practised in food industries, the mechanism of fracture is still not completely understood. In the process, a mechanical component stresses the materials by mechanical action of the grinding machine. Initially, the stress is internally absorbed by the material as strain energy. Fracture occurs along the lines of weakness when the local strain energy exceeds the critical level. It is a function of the material. Fracture releases stored energy and the energy is used in the creation of new surfaces, but the major part of it is dissipated as heat. The material may fracture even at low stress concentrations when maintained for a longer period. Hence, grinding is achieved by mechanical stress followed by rupture. The hardness of the material and its tendency to crack decides the energy requirement for fracturing and size reduction processes. The factors that affect the grinding process are the applied force (which may be compression, impact or shear), the duration of the force application and the magnitude of the applied force.

A grinding process will be efficient when the energy applied to the material is marginally higher than the energy needed to rupture the material. Excess energy is lost in the form of heat and this loss should be as low as possible. The grinding process is thus characterized by the amount of energy used and the magnitude of the new surface formed.

2.2 Applications of the grinding process

All three classical laws of grinding (Kick's, Rittinger's and Bond's laws) seem to be applicable although Rittinger's law shows better suitability than the other two, followed by Bond's law for use with various wet grinding systems like the mixer grinder, stone grinder and colloid mill. Predominant compressive forces involved in stone grinding are reflected by higher starch damage in batter, which is also evident in the photomicrographs (Sharma *et al.*, 2008).

2.2.1 Dry grinding

The energy required for single-stage size reduction is higher when compared to a multistage size reduction (Dziki, 2011). The specific energy required to grind a whole wheat kernel is 32.6–79.1 kJ/kg in the single stage while it is 23.1–44.4 kJ/kg for the two-stage process.

Recovery of protein from unclassified primary ground flour of soybean and okara is much lower compared to their corresponding fine fractions (particle size about 75 μm) (Vishwanathan, Singh and Subramanian, 2011a). Secondary grinding of a coarse fraction improves the overall protein recovery to an extent of 3.3% for okara and to a much larger extent of 6.8% for soybean.

High-energy ball milling has been reported to help in making mesoporous materials from egg shell (Tsai *et al.*, 2008). The microwave dried samples have been ground in a hammer mill and Bond's work index is found to decrease with an increase in drying time (Velu *et al.*, 2006). Microwave drying prior to grinding facilitates in reducing energy consumption. Bond's work index for microwave-dried gum samples is 0.48 kWh/kg compared to 0.76 kWh/kg for the control sample having the same moisture content (Walde *et al.*, 1997). The energy required for grinding dry carrot grits increases as the moisture content is increased from 10 to 15%; however, it decreases as the moisture content is increased to 18% and, again, shows an increasing trend. A moisture content of 18% has been recommended for minimal energy for grinding (Chakkaravarthi *et al.*, 1993).

Grinding of split legumes to initial coarse grinding and then to fine particles has been studied by Indira and Bhattacharya (2006). Among various legumes, lentil (*Lens esculenta*) is the most amenable to grinding, resulting in the creation of very high surface areas. In contrast, under the same conditions, Bengalgram (*Cicer arietinum*) offers the least surface area. Cowpea (*Vigna unguiculata*), blackgram (*Phaseolus mungo*) and greengram (*Phaseolus aureus*) have similar grinding characteristics. From particle size distribution, it is evident that grinding characteristics of legumes largely depend on an inherent property and not merely on the size or shape of the samples.

The sprouting process has a significant influence on grinding characteristics of wheat (Dziki and Laskowski, 2010). The average particle size of the sprouted kernels is significantly lower than those from the matured whole kernels. Sprouting causes an increase in the fraction of particles below the 200 μm size and a decrease occurs in the value of specific grinding energy. Specific grinding energy ranges from 35.5 to 141.6 kJ/kg and from 41.4 to 164.3 kJ/kg for the sprouted and matured whole kernels, respectively.

Muller (2003) has made an effort to correlate the noise made by a mill when grinding malt to its friability assessment using computer-based digital sample analysis. The intensity of noise generated by a laboratory disc mill shows an inverse effect on the friability. In the case of wheat farina, Greffeuille *et al.* (2007) have reported that cultivars (cultivated variety) possess an important

role in particle size distribution of the milled products and the energy required for milling. Both grain hardness and vitreousness affect the grinding index (energy required to produce 1 kg of flour). In soft wheat grains, phospholipids associated with friable parts and glycolipid-enriched parts offer high resistances to grinding. While grinding pea seeds, the chemical and physical characteristics of the size fractions of hulls and kernels seem to have an effect on different comminution laws (Maaroufi *et al.*, 2000). This phenomenon has led to a physical separation of botanical constituents like hulls and parietal constituents in coarse fractions, kernel and cellular constituents to finer fractions and starch granules in the smallest fractions.

Gaete-Garretón *et al.* (2003) have reported that ultrasound enhances the performance of a roller mill. The energy consumption for grinding appears to be significantly reduced by careful application of an ultrasonic field in the grinding zone. Also, the use of ultrasound in the grinding zone lowers stress on the shafts and reduces the torque required. There is a reduction in abrasive wear of the rolls due to lower mechanical stress. Ozone treatments of wheat grains before milling is reported to significantly reduce (10–20%) the energy required without affecting the flour yield (Desvignes *et al.*, 2008). The resistance offered by wheat endosperm to rupture is affected by a reduction of the aleurone layer extensibility due to ozone treatment, which may explain the reason for the reduction in milling energy.

2.2.2 Wet grinding

Wet grinding of hydrated soybean has been explored using various grinding systems (Vishwanathan *et al.*, 2011a, 2011b). Greater particle size reduction is observed by using a mixer grinder; this is more significant since the protein recovery depends on particle size. In respect of energy consumption, the colloid mill is the best. Advantages of continuous operation and energy conservation make the colloid mill a preferred system for industrial-scale operation. The colloid mill (Solanki *et al.*, 2005) has the potential to be used for wet grinding of rice and blackgram on an industrial scale for the manufacturers of different traditional Indian foods. The colloid mill can ensure the required starch damage during wet grinding, which is essential for proper fermentation of batter during the preparation of traditional products such as *idli* and *dosa*.

Various wet grinding systems like the mixer grinder, stone grinder and colloid mill have been evaluated for wet grinding of raw and parboiled rice (Sharma *et al.*, 2008). In batch grinders, the duration of grinding has an inverse effect on the final particle size and direct impact on starch damage as well as energy consumption. The stone grinder is less energy efficient than the mixer grinder during size reduction of raw rice. Parboiled rice needs a longer grinding duration when compared to raw rice. The stone grinder predominantly employs compressive forces for grinding, which is evident in

higher starch damage in batter, which is also confirmed by photomicrographs. Parboiled rice slurry is more viscous than the raw rice sample.

Pan and Tangratanavalee (2003) reported that solid loss increases significantly as the soaking temperature is increased from 30 to 40 °C. Soaking conditions do not affect the grinding property while the final moisture content affects grinding characteristics. High-temperature soaking significantly reduces the required soaking time. The optimum level of moisture content for better wet grinding properties is 120% (dry basis).

2.3 Grinding energy laws

The exact power requirement for specific size reduction is difficult to determine. The type of material, moisture content, fineness of grinding, rate of feed, type and condition of mill, etc., affect the power requirement. However, the energy laws may help in a relative comparison for optimizing the process variables. There are some theories with an assumption to calculate the energy required for grinding. The assumption is that the energy required is the simple power function of L (Earle and Earle, 2006):

$$\mathrm{d}E/\mathrm{d}L = KL^n \tag{2.1}$$

where $\mathrm{d}E$ is the differential grinding energy, $\mathrm{d}L$ is the typical change in dimension, L is the typical length dimension and K and n are constants.

Several energy laws have been proposed to relate size reduction to a single variable, i.e. the energy input to the mill. These laws are:

Kick′s law (when $n = -1$)	$E = K_{\mathrm{K}} f_{\mathrm{c}} \ln(L_1/L_2)$	(2.2)
Rittinger′s law (when $n = -2$)	$E = K_{\mathrm{R}} f_{\mathrm{c}} (1/L_2 - 1/L_1)$	(2.3)
Bond′s work index (when $n = -1.5$)	$E = E_i (100/L_2)^{1/2}[1 - (1/q)^{1/2}]$	(2.4)

where f_{c} is crushing strength, K_{K} is Kick's constant, K_{R} is Rittinger's constant, E_i is Bond's work index, $q = L_1/L_2$ and L_1 and L_2 are the initial and final particle sizes, respectively.

None of the energy laws apply well in practice (Perry and Green, 1997). The main use of these laws is in making comparisons between energy requirements for various degrees of size reduction. Bond's work index is more realistic in depicting the performance of commercial crushers and grinders (McCabe, Smith and Horriott, 1993). Tables 2.1 and 2.2 show the energy-related indices for different dry and wet grinding processes.

Table 2.1 Bond's work index for various food ingredients

Food ingredient (dry grinding)	Pre-treatment	Bond's work index (kW h/kg)	Reference
Wheat	Microwave dried	2.41	Walde *et al.* (2002)
Carrot grits	Conventionally dried	90.19	Chakkaravarthi *et al.* (1993)
Gum Karaya	Microwave dried	0.36–0.52	Walde *et al.* (1997)
Gum Karaya	Conventionally dried	0.76	Walde *et al.* (1997)
Cumin	Cryogenic grinding	0.053–0.086	Goswami and Singh (2003)
Maize	Microwave dried	0.08–0.28	Velu *et al.* (2006)
Coconut residue	–	0.07	Raghavendra *et al.* (2006)

Table 2.2 Energy expenditure for different wet grinding processes

Ingredient	Grinding system	Energy expenditure (kJ/kg)	Reference
Rice (raw)	Stone grinder	22.5	Sharma *et al.* (2008)
	Mixer grinder	10.8	
	Colloid mill	12.1	
Rice (parboiled)	Stone grinder	32.7	
	Mixer grinder	33.0	
	Colloid mill	38.4	
Soy bean	Mixer grinder	132.8	Vishwanathan, Singh and Subramanian (2011a, 2011b)

Morrell (2004) has proposed an alternate energy–particle size relationship in a similar approach to Bond's work index. The proposed relationship for milling in a ball mill is comparable to those obtained using Bond's equation and can offer a more precise prediction of specific power requirements.

2.4 Machinery requirement

Based on the breaking action, size reduction equipments are divided into two classes: crushers and grinders. Crushers work on the principle of compression and are used in heavy industries, whereas grinders work on the principle of shear and impact combined with compression. Even uniform sized particles are subjected to size reduction and the size of the ground particles produced will vary from coarse to fine and even to dust. Further on in the process, the coarser particles are further reduced while finer particles remain less affected.

The principal types of size-reduction machines are as follows:

A. Crushers (coarse and fine)
   1. Jaw crushers
   2. Gyratory crushers
   3. Crushing rolls

B. Grinders (intermediate and fine)
   1. Hammer mills and impactors
   2. Rolling–compression mills
   3. Attrition mills
   4. Tumbling mills

C. Ultrafine grinders
   1. Hammer mills with internal classification
   2. Fluid–energy mills
   3. Agitated mills
   4. Ball mills

D. Cutting machines
   1. Knife cutters, dicers and slitters

2.4.1 Crushers

Jaw and gyratory crushers draw material down into a progressively narrower space resulting in size reduction. However, they are not extensively used in the food industries.

2.4.2 Grinders

There are various types of grinders. These are hammer mill, plate/disc mill, pin mill, roller mill, etc.

Hammer mill A rotor is attached with swinging or stationary hammer heads, which rotate at a high speed to force the ingredients against a circular screen or a solid serrated section, resulting in crushing of material inside a hardened casing (Figure 2.1). The crushed materials are further crushed until they reach the size of the aperture of a hard metal sieve at the outlet. These mills can handle both brittle and fibrous dry materials. Hammer mills can grind ingredients up to a maximum size of 75 mm, while larger pieces above 75 mm require a pre-crusher for initial size reduction.

Two types of arrangement exist in the hammer mill; the common one is a horizontal drive shaft, which suspends vertical hammers to crush any friable and fibrous dry materials containing less fat. The other one is the vertical

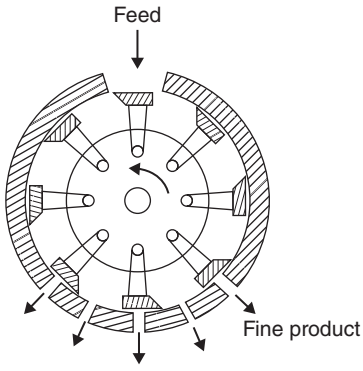

Figure 2.1 Hammer mill (source: R.L Earle and M.D. Earle. Reproduced with permission of The New Zealand Institute of Food Science & Technology.)

hammer mill where the drive shaft is positioned vertically while screens and hammers are positioned horizontally. Materials successfully reach the screen hole size and are carried by the gravity force outside the mill and then by air or conveyor to storage. Bigger particles, which still escape the size reduction process, are allowed to drop through the mill and may be recycled for size reduction. This facilitates the separation of foreign materials, such as metal and stones, which can cause screen damage.

The important factors affecting the grinding capacity are:

- Number of hammers on a rotating shaft
- Speed of rotation
- Hammer size
- Arrangement of hammers
- Sharpness
- Wear patterns and
- Clearance between the tip and screen or striking plate.

The residence time of material inside the grinder and air flow characteristics in the grinding zone are related to heat generated during the size reduction process.

Attrition mill Attrition mills are also known as plate mills (Figure 2.2) or disc pulverizes and are widely used for small-scale milling. These mills use the working principle of a hammer mill of impact to a certain extent. However, they also impart the shearing and cutting actions. The material is fed in between two circular plates with the flute or roughened surface. One of the plates is fixed while the other one has a rotation facility. Normally, the material is fed near the axis of the rotation and is sheared and crushed as it makes its

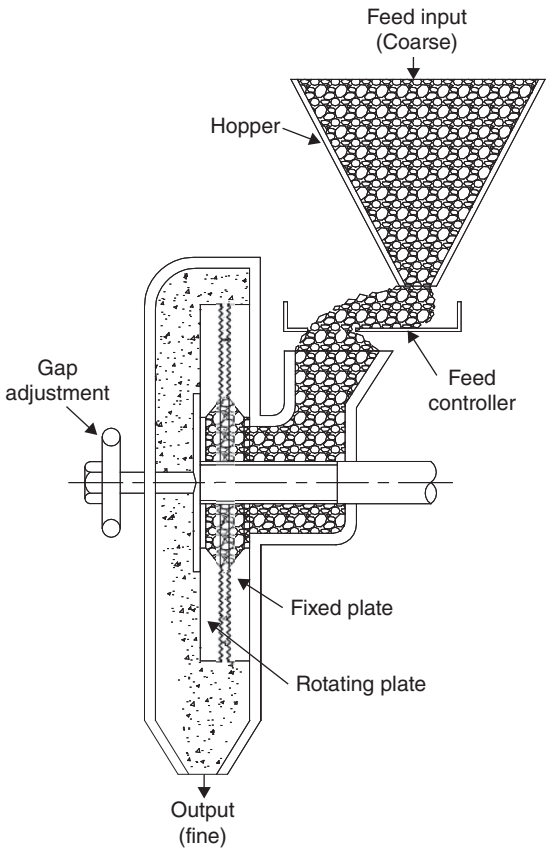

Figure 2.2 Attrition mill

way out to the edge of the plate. These mills produce a narrow range of particle sizes. The low clearance and higher speed facilitate the production of finer size particles. The plate mills led to the development of the colloid mill. The main difference between them is clearance between the plates and the speed of rotation. These mills are extremely versatile and can be designed to varied end purposes like shred, curl, granulate, grind, shear, twist, blend, rub, fibreize, pulverize, crack, cut, fluff, hull, refine, etc. The various plate designs/styles are shown in Table 2.3.

Pin mill These mills have two discs with orderly placed pins to impart high-impact and shear forces for size reduction. One disc is the stator disc and the other is the rotary disc (Figure 2.3). A consistent and uniform quantity of material is gravity fed through the centrally located inlet of the stator disc. Centrifugal forces accelerate the materials in such a way that it reaches the impact zone. Control of the rotor speed helps to achieve the desired

Table 2.3 Different plate mills and their characteristics

Type of design	Characteristics
Spiral rib plate	They are used for general purpose grinding and are versatile for varied applications
Radial rib plate	Applied for refining the moist/sticky materials to milling and hulling of food products and cellulose. These machines are available in coarse, medium and fine configurations for different applications
Oval rib plate	Specialized designs are available for curling
Radial undercut rib plate	Mainly used for high volume grain, corn cob and other agricultural oriented milling needs at a low cost of processing

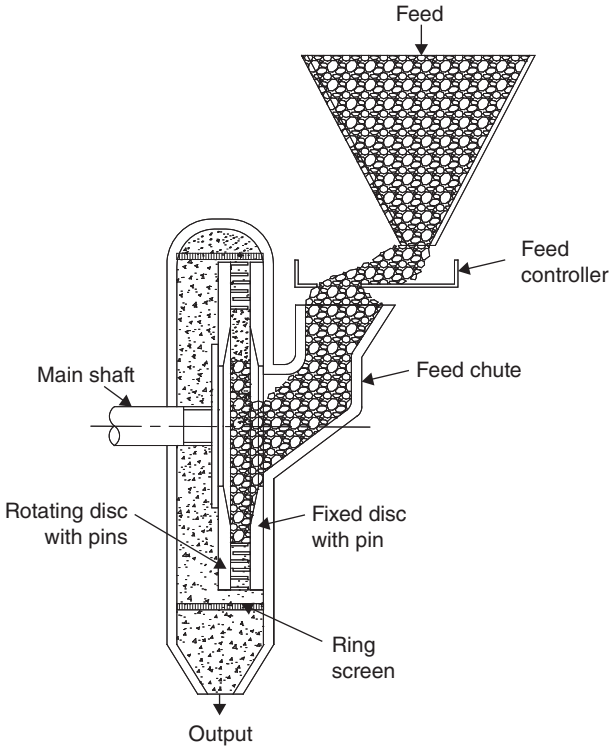

Figure 2.3 Pin mill

narrow spectrum of particle size distribution. Variation in the speed of rotor facilitates the mills to use it for coarse grinding or de-agglomeration as well as for fine grinding.

Roller mill Smooth or finely fluted rolls rotate at the same speed as for normal milling and at different speeds for tearing action. These types of mill (Figure

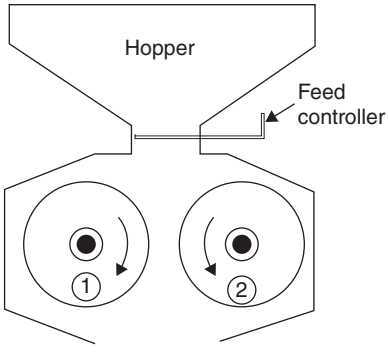

Figure 2.4 Schematic view of a roller mill: 1. adjustable roller and 2. fixed roller

2.4) are widely used in industries to grind grains like corn (maize), wheat, rye, barley or malt into flour. The larger surface area in grinding allows more material to be ground quickly. The narrow particle size distribution, minimal dust in the absence of high-speed impact, low temperature rise in the absence of product recirculation and low energy consumption are the major advantages of these types of mill. A combination of cutting, attrition and crushing occurs in roller mills. Roller mill size reduction is economical but limited to materials that are fairly dry and low in fat.

2.5 Mechanism of size reduction

Most of the food ingredients are brought for size reduction after the unit operations like grading, pre-cleaning, drying, etc. These unit operations induce internal stress and initial flaws of different degrees. Flaws are the weakest point in a particle and facilitate size reduction (Figure 2.5). During the size

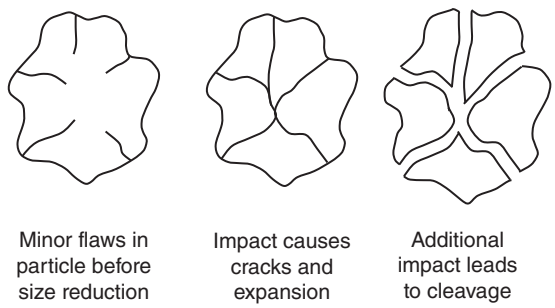

Figure 2.5 Flaws leading to cleavage of materials during processing

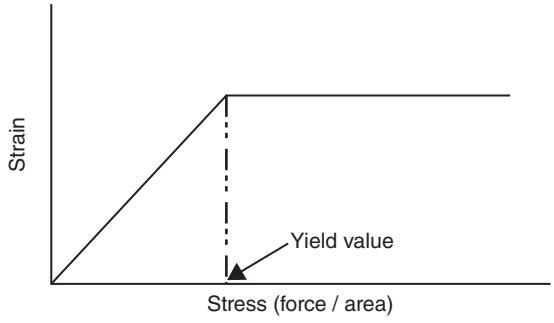

Figure 2.6 Stress–strain relationship of materials

reduction process, the food ingredients are subjected to one or a combination of basic forces like compression (crushing force), shear (cutting force) and tension (elongating/pulling force). These forces increase flaws to a critical value, which is called the yield value, and the material starts breaking down. The stress–strain relationship (Figure 2.6) indicates that an increase in stress increases strain, leading to a yield value and subsequent breakage.

2.5.1 Grinding of heat-sensitive and fat-containing materials

Heat is generated during milling due to the long stay of material inside the grinding zone, that is until it reaches the aperture openings of a screen fixed in the delivery side. Such heat can melt heat-sensitive materials like fat, result in the loss of heat-sensitive nutrients like vitamins, nutraceuticals and bioactive components. The solution to this heat generation is cooling the grinding chamber surfaces and/or cooling the material prior to milling. This can be achieved by water-jacketing the grinding zone, injecting cryogen like liquid nitrogen into the grinding zone and/or exposing the feed to cryogen prior to milling.

Semi-solid materials or fatty materials like cheese, pepper sauce, baby food, etc., can pose extreme difficulty in grinding, because these materials can stick to or get trapped in the screen section of mills and can block the screen apertures. These materials can also stick to the surfaces of the grinding chamber. This problem can be solved by a custom-designed milling system, which facilitates positive movement of the ground material from the grinding zone without sticking or clogging. Alternatively, internal air jet or water injectors can be used to ensure material movement from the grinding zone to the discharge section. A high-speed impeller with a specialized design ensures that air currents throw particles away from the grinding zone. Further, the air current pushes out all the material to exit and ensures cleanliness. Air flow also helps to minimize heat generation and in turn reduces heat-induced damages.

Table 2.4 Different cutting operations

Cutting operations	Description
Slicing	Blade slices by centrifugal force and each slice falls away freely
Dicing	Vegetables, fruits and meats are first sliced and then cut into strips by rotating blades. The strips are then fed to a second set of rotating knives that operate at right angles to the first set and cut the strips into cubes
Shredding	Commonly they are the modified form of a hammer mill in which knives are used instead of hammers to produce a cutting action. A second type of shredder, known as the squirrel cage disintegrator, has two concentric cylindrical cages inside a casing. They are fitted with knife blades along their length and the two cages rotate in opposite directions. Food is subjected to powerful shearing and cutting forces as it passes between them
Pulping	A combination of compression and shearing forces are used for juice/pulp extraction from fruits or vegetables and for producing pureed and pulped meats

2.5.2 Cutting of fruits and vegetables

Size reduction of fresh produces like fruits and vegetables before the end use is normally done by chopping, slicing, shredding, peeling, dicing and sectioning (Fellows, 2000). Irrespective of the method employed for cutting, the produce must be uniform in size as nonuniform sized products are not appealing to consumers. This type of size reduction can be classified in order of decreasing particle size (Table 2.4).

2.6 Size reduction of liquid

Size reduction of liquid is done by homogenization and atomization and is mostly targeted for stabilization of the sample and/or for further processing. In this process, size reduction and dispersion of the solid, liquid or gas phase is carried out within a continuous phase, which is different from the phase that is dispersed. Most of the time the fat globules in the formulation are reduced in size and dispersed uniformly such that the formulation is stable against creaming (Fellows, 2000). Wet grinding produces fine particles directly in a liquid phase.

2.6.1 Homogenization

The unit operation that prevents fat globules from coalescing into cream is called homogenization, which is also a way of size reduction in liquids. The liquid formulation is forced through a small opening at higher speeds for

breaking down the fat or other globules into smaller ones. The five main types of homogenizer are as follows.

High-speed mixers Edges and tips of the blades in high-speed turbine/propeller type mixers impart a shearing action on the low viscous food formulations to homogenize into a smooth homogeneous emulsion.

Pressure homogenizers Pressure homogenization is conventionally done prior to pasteurization and ultra-high-temperature (UHT) sterilization. Pressure homogenizers use a high pressure pump, operating at 100–700 bar, which is fitted with a homogenizing valve(s) (two-stage homogenization) on the discharge side. When liquid is pumped through the small adjustable gap (< 300 μm) between the valve and the valve seat, the high pressure produces a high liquid velocity (80–150 m/s). An instantaneous drop in velocity occurs as the liquid emerges from the valve. This extreme turbulence produces powerful shearing forces and the droplets in the dispersed phase become disrupted. The collapse of air bubbles (termed 'cavitation') and impact forces created in some valves by placing a hard surface (a breaker ring) in the path of the liquid further reduces the globule size.

Colloid mills Colloid mills are more effective than pressure homogenizers in creating high shear and are meant for high viscous liquids. They are essentially vertical disc mills with a narrow gap between stationary and rotating discs in the range of 0.05–1.3 mm and rotate at 3000–15000 rpm. Numerous designs of disc including flat, corrugated, conical shapes and even carborundum are available for different applications. The greater friction created during size reduction of high-viscous foods may require these mills to be cooled by circulating water in the water jacket.

Ultrasonic homogenizers A high-frequency sound wave in the range of 18–30 kHz is used in ultrasonic homogenizers to cause alternate cycles of compression and tension in low-viscosity liquids. It is also responsible for cavitation of air bubbles to form emulsions with droplet sizes of 1–2 μm. This type of homogenizer is used for the production of salad creams, ice cream, synthetic creams, baby foods and essential oil emulsions. It is also used for dispersing powders in liquids.

Hydro-shear homogenizers and microfluidizers In a hydro-shear homogenizer, the feed liquid enters the chamber at a high velocity and is made to spin in increasingly smaller circles and increasing velocity until it reaches the centre and is then discharged. The differences in velocity between adjacent layers of liquid causes high shearing forces, which, together with cavitation

and ultra-high-frequency vibration, break droplets in the dispersed phase and results in stable emulsion.

2.6.2 Atomization

Atomization is a process of transforming the bulk of a liquid into fine droplets. The size of the droplets, when sprayed in vacuum or in gas, varies from submicrometres to several hundred micrometres in diameter. Numerous spray devices like atomizers, nozzles and applicators are used for size reduction. In all these devices, atomization is achieved by creating a high velocity between the liquid and the surrounding gas (air). A spray is a system of droplets immersed in the gaseous continuous phase. Atomization is widely used in spray drying and surface coating of food products and is also a size reduction process to increase the surface area so as to facilitate further unit operations like spray drying, evaporative cooling, encapsulation and spray coating.

2.7 Conclusions

Even though there are exhaustive developments in the area of dry grinding, the process of wet grinding has been left unattended as only a few published papers are available. A large-scale wet grinding system used in mineral industries possesses limitations for scaling down to suit the food processing sector. Continuous wet grinding is an urgent need of food processing since wet grinding can minimize the loss of thermally sensitive constituents in food. A systematic effort in developing a continuous medium-scale wet grinding system is the need of the hour and such a system will help the traditional food sector. An energy-efficient grinding system for both dry grinding and wet grinding deserves research efforts since grinding is an energy-intensive unit operation. Development of appropriate models for predicting energy requirements for size reduction of specific ingredients by incorporating material properties like hardness and brittleness apart from particle size will be a greater challenge.

References

Chakkaravarthi, A., Math, R.G., Walde, S.G. and Rao, D.G. (1993) Grinding characteristics of carrots (*Daucus carota* L.), *Journal of Food Engineering*, **20**, 381–389.

Desvignes, C., Chaurand, M., Dubois, M., Sadoudi, A., Abecassis, J. and Lullien-Pellerin, V. (2008) Changes in common wheat grain milling behavior and tissue mechanical properties following ozone treatment. *Journal of Cereal Science*, **47**, 245–251.

Dziki, D. (2011) Effect of preliminary grinding of the wheat grain on the pulverizing process. *Journal of Food Engineering*, **104**, 585–591.

Dziki, D. and Laskowski, J. (2010) Study to analyze the influence of sprouting of the wheat grain on the grinding process. *Journal of Food Engineering*, **96**, 562–567.

Earle, R.L. and Earle, M.D. (1996) Unit Operations in Food Processing, Web edition. The New Zealand Institute of Food Science and Technology (Inc.), available in http://www.nzifst.org.nz/unitoperations/sizereduction1.htm#energy. Assessed in August 2013.

Fellows, P. (2000) *Food Processing Technology*, Woodhead Publishing Limited, Cambridge, England, pp. 103–115.

Gaete-Garretón, L., Vargas-Hernandez, Y., Chamayou, A., Dodds, J.A., Valderama-Reyes, W. and Montoya-Vitini, F. (2003) Development of an ultrasonic high-pressure roller press. *Chemical Engineering Science*, **58**, 4317–4322.

Goswami, T.K. and Singh, M. (2003) Role of feed rate and temperature in attrition grinding of cumin. *Journal of Food Engineering*, **59**, 285–290.

Greffeuille, V., Abecassis, J., Barouh, N., Villeneuve, P., Mabille, F., Bar L'Helgouac'h, C. and Lullien-Pellerin, V. (2007) Analysis of the milling reduction of bread wheat farina: physical and biochemical characterization. *Journal of Cereal Science*, **45**, 97–105.

Indira, T.N. and Bhattacharya, S. (2006) Grinding characteristics of some legumes. *Journal of Food Engineering*, **76**, 113–118.

Maaroufi, C., Melcion, J.-P., de Monredon, F., Giboulot, B. and Guibert, D. (2000) Marie-Pierre Le Guen, fractionation of pea flour with pilot scale sieving. I. Physical and chemical characteristics of pea seed fractions. *Animal Feed Science and Technology*, **85**, 61–78.

McCabe, W.Z., Smith, I.C. and Horriott, P. (1993) *Unit Operations of Chemical Engineering*, 5th edn. McGraw-Hill, New York, pp. 962–964.

Morrell, S. (2004) An alternative energy–size relationship to that proposed by Bond for the design and optimisation of grinding circuits. *International Journal of Mineral Processing*, **74**, 133–141.

Muller, R. (2003) An assessment of malt friability from analysis of the sound made during milling. *Journal of Cereal Science*, **37**, 241–251.

Pan, Z. and Tangratanavalee, W. (2003) Characteristics of soybeans as affected by soaking conditions. *LWT - Food Science and Technology*, **36**, 143–151.

Perry, R.H. and Green, D.W. (1997) *Perry's Chemical Engineers' Handbook*, 7th edn, Chapter 20. McGraw-Hill, New York, p. 13.

Raghavendra, S.N., Ramachandra Swamy, S.R., Rastogi, N.K., Raghavarao, K.S.M.S., Kumar, S. and Tharanathan, R.N. (2006) Grinding characteristics and hydration properties of coconut residue: a source of dietary fiber. *Journal of Food Engineering*, **72**, 281–286.

Sharma, P., Chakkaravarthi, A., Singh, V. and Subramanian, R. (2008) Grinding characteristics and batter quality of rice in different wet grinding systems. *Journal of Food Engineering*, **88**, 499–506.

Solanki, S.N., Subramanian, R., Singh, V., Ali, S.Z. and Manohar, B. (2005) Scope of colloid mill for industrial wet grinding for batter preparation of some Indian snack foods. *Journal of Food Engineering*, **69**, 23–30.

Tsai, Wen-Tien, Yang, Jwu-Maw, Hsu, Hsin-Chieh, Lin, Chien-Ming, Lin, Keng-Yu and Chiu, Chun-Hsiang (2008) Development and characterization of mesoporosity in eggshell ground by planetary ball milling. *Microporous and Mesoporous Materials*, **111**, 379–386.

Velu, V., Nagender, A., Prabhakara Rao, P.G. and Rao, D.G. (2006) Dry milling characteristics of microwave dried maize grains (*Zea mays* L.). *Journal of Food Engineering*, **74**, 30–36.

Vishwanathan, K.H., Singh, V. and Subramanian, R. (2011a) Influence of particle size on protein extractability from soybean and okara. *Journal of Food Engineering*, **102**, 240–246.

Vishwanathan, K.H., Singh, V. and Subramanian, R. (2011b) Wet grinding characteristics of soybean for soymilk extraction. *Journal of Food Engineering*, **106**, 28–34.

Walde, S.G., Balaswamy, K., Shivaswamy, R., Chakkaravarthi, A. and Rao, D.G. (1997) Microwave drying and grinding characteristics of gum karaya (*Sterculia urens*). *Journal of Food Engineering*, **31**, 305–313.

Walde, S.G., Balaswamy, K., Velu, V. and Rao, D.G. (2002) Microwave drying and grinding characteristics of wheat (*Triticum aestivum*). *Journal of Food Engineering*, **55**, 271–276.

3

Dough Processing: Sheeting, Shaping, Flattening and Rolling

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3.1 Introduction

Baked goods are available in a variety of compositions, shapes, and textures, which are the determining characteristics leading to consumer purchase. Forming or shaping of the product during manufacturing involves multiple unit operations such as rolling, sheeting, and flattening, which are performed with the aim of attaining the desired portion size of dough, which can be finally shaped using a shaping or forming operation. Depending on the type of dough, shaping can be as simple as the stamping of a mold on a thin layer of dough (e.g., crackers and biscuits) or a complicated operation like, for example, the ones used for leavened breads and other products such as croissants and filled pastries.

For typical solids, that is, elastic materials, these processes are well defined, and conditions such as temperature and pressure can be adjusted to improve the material malleability/ductility and make them conducive to the process. However, doughs are viscoelastic materials that exhibit normal stresses, stress generation, and relaxation, and also strain recovery effects during the many types of deformation that are prevalent in dough processing operations. Thus, the complexity of the material behavior during processing makes forming and shaping operations more difficult to control. In addition to the application of

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stress or deformations, dough processing includes resting steps to allow for stress relaxation during the shaping of the dough material. It is hypothesized that if a suitable relaxation time is used during the shaping opreation to permit the release of the internal stresses generated in the dough during the process, a better control can be achieved over the final shape of baked goods.

The chemical composition and structure of the dough and how its protein component reacts to the applied stress determines the choice of technology to be used in the process. For instance, corn masa, a type of dough used in the production of corn tortilla and corn chips, exhibits predominantly a viscous behavior with little elasticity; thus it can be sheeted in a single pass. Conversely, wheat flour dough is more viscoelastic in nature due to the development of its gluten proteins. The typical viscoelastic behavior of wheat flour dough can be described as a viscous flow under stress, but as soon as the stress is released the dough tends to revert back partially to the starting shape due to elasticity, a phenomenon known as strain recovery. In addition, it has been well documented that any work in the form of mixing or any other type of deformation that is applied to dough, is used for transformation of gluten proteins, which contribute to the development of the dough (Zhen *et al.*, 2000). Some of that energy also results in the generation of internal stress in the dough, which must be dissipated by allowing for a proper relaxing time so that the formed pieces in the shaping operation may retain the final shape.

Among the different operations used in dough processing, it is found that a few are very relevant and have a large impact on the quality of the final product. They, among others, include sheeting, shaping, and rolling.

3.2 Dough sheeting

3.2.1 Technology

Sheeting is a process that was initiated to process plastics, where it is also known as calendering. Calendering is an important process for the formation of plastic sheets that have been extensively studied and there exist reports in the academic and industrial arena describing and modeling it. Although the technology of plastic calendering and dough sheeting differ in many aspects, principally on the type of the material being processed, there is a basic common approach to study and model the process. In addition to the inherent different characteristics on the rheology of plastics and dough, calendering is generally performed at high temperatures whereas dough is processed generally at low or room temperatures. The condition of elevated temperature makes the processed plastic behave as a free-flowing fluid material. Conversely, a dough is generally processed below or at room temperatures where it is a semi-liquid viscoelastic material. Plastics usually have a homogeneous composition formed by macromolecules having narrow molecular size whereas the composition of dough is far from being

homogeneous and is composed of macromolecules having large molecular size distribution. Despite the significant differences in the rheology of the material being processed, in essence both processes are similar in various technological aspects. For instance, both are based on the passing of the material through rotating rolls and, although the characteristics of the raw materials differ significantly, on certain occasions it is worth looking at the literature on calendering, which for reasons of space is not reviewed in this chapter, but for further information the reader could refer to two excellent and complete references (Pearson, 1985; Middleman, 1977). Instead the chapter is devoted solely to the processing of dough.

Dough sheeting is an indispensable component in the processing of many baked products. It provides the final shape and structural identity of the baked product, irrespective of the type of dough, before it goes to the baking oven. Most of the baked products can be classified on the basis of the type of dough. Detailed differences on the types of dough used in the baking industry are covered in other parts of this book (e.g., refer to the dough mixing section). Depending on the flour used in its preparation, moisture content, and the addition of other ingredients, doughs are commonly classified as either soft or hard doughs. In the case of soft dough, water and flour are the major ingredients, which after proper formulation and upon mixing result in a cohesive mass due to the development of the gluten proteins. Dough prepared in this way is often referred to as being low-fat and low-sugar formulations. Most of the soft dough formulated with low content of fat and sugar exhibits a dominant elastic behavior, which plays a vital role in determining the type of sheeting, molding, and flattening operations to be performed in the process. Typical examples are doughs used for baked goods such as bread, pizza, and buns.

The other type of dough formulation includes that having high fat and sugar content to produce the so-called hard dough. For baked products produced with high fat and high sugar formulations, the formation of a gluten network is not desired resulting in a dough mass having lower cohesiveness. Due to the undeveloped gluten network and the absence of a high elastic component, these doughs exhibit plastic-like behavior. Doughs with a plastic behaviour do not regain their original shape once the stress acting on the dough is removed, that is, they can retain the shape of the mold easily during the molding operation. Typical examples of these doughs are those used in the preparation of baked goods such as biscuits and cookies.

Once the dough is mixed, with the aim of achieving a relatively uniform distribution of the ingredients, it is dumped into a hopper where the sheeting operation begins. Sometimes and for small-scale operations a cutting step to obtain small pieces of dough that can be handled manually is included, for example, for the production of artisan pastry making. However, most industrial sheeting machines forming part of a continuous production line compress the irregular piece of dough coming from the hopper between two rotating rolls, resulting in the formation of a relatively thick sheet with uniform edges

and thickness, and having little or no holes. This sheet of dough is then passed repeatedly through multiple sets of smooth rolls rotating either in opposite directions with the same speed or in the same direction with a speed differential. The gap between each set of rollers is gradually reduced to produce thin smooth sheets at the end of the process line, which can be then molded or cut into desired shapes before baking. There are many advantages obtained from the sheeting operations and one of the most relevant reported is the improvement in the final baked good crumb texture, which is the major contributing factor to the baked product quality. Various hypotheses have been proposed to explain this quality improvement and are discussed by Kilborn and Tipples (1974).

Sheeting of dough serves multiple purposes; the main one is the conversion of a dough mass of irregular shape coming out from the mixer into a sheet with an even thickness covering a large part of the width of the conveyor in the baking line. The first sheet of dough formed, typically, has a uniform thickness and smooth edges with absence of breakages or holes. The latter is important to ensure a uniform cohesive sheet with no cracks or holes when the dough is recycled from the line and re-mixed. The type of dough sought determines the number of passes necessary to attain the final dough thickness.

Depending on the sheeting system, the sheeting operation can be classified by various manners. For instance, in the classical sheeting pin, the dough is passed through one sheeting surface and one stationary surface. Conversely, various configurations can be possible with two moving surfaces, especially for mechanized systems where two sheeting surfaces are operating either at the same speed or with a speed differential. For the case of two rollers operating at different speeds, the roller operating at the faster speed stretches the dough relative to the slower roller, leading to a better size reduction as well as alignment of the gluten network. Thus, the desired size reduction or sheeting can be achieved in a relatively smaller number of passes, which means savings in terms of space and energy consumption.

Another major purpose of the dough sheeting operation, in addition to the thickness size reduction, is the development of the gluten network. Gluten network development is a major contributor to the final quality of bread, especially its crumb quality. Typically during sheeting, the dough is passed between two surfaces, both of which are moving with either different speeds or one is stationary. As the dough passes through the sheeting station it experiences input of energy as well as a squeezing action (Moss, 1980; Stenvert *et al.*, 1979) resulting in improvement of the bread crumb structure. Kilborn and Tipples (1974) measured the power consumption of rollers during sheeting, in addition to bread crumb quality, and observed that the mixing and stretching of the dough during sheeting is not only highly appropriate for good product quality but also results in an operation that is more energy efficient than mixing. Typical dough development using sheeting would need about 10 to 15% of the energy needed to develop the dough at the same extent in a conventional

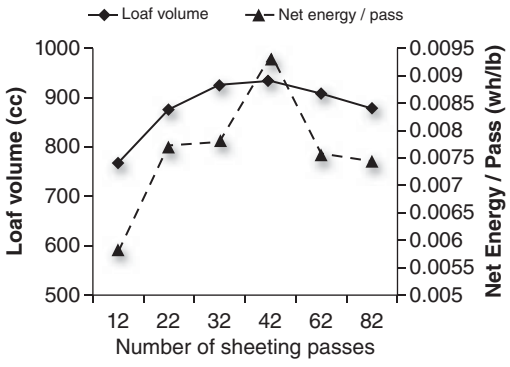

Figure 3.1 Effect of the number of sheeting of passes on net energy per pass and loaf volumes. (The figure is based on data tabulated by Kilborn and Tipples (1974). One sheeting pass is defined as a pass between two pairs of sheeting rolls with a gap of 7/32 in at the first pair of rolls followed by a second pair of sheeting rolls with a gap of 5/32 in. The resulting dough sheet was folded in half and turned 90° for the next pass.)

mixing process. Data on baked loaf volume have shown that the baked volume reaches a peak at 42 sheeting passes and the energy required for sheeting also peaked at the same number of passes (Figure 3.1). This behaviour is similar to that observed on the mixing of dough where the power during dough development in different mixers also peaks at the maximum of dough development, measured by a number of rheological techniques (Zheng *et al.*, 2000). In other words, dough exhibits optimum gluten network development at the highest power input; overmixing leads to breakdown of the gluten network, resulting in a lower power input and lower baked loaf volumes. The proved advantage of mixing and dough development produced by the sheeting process makes the process suitable for the production of no-time dough, where high levels of oxidative agents are used. Sheeting has been also practiced for the production of bread in different parts of the world, such as Spain, South America, Southeast Asia, and Africa (Stenvert *et al.*, 1979). However, this advantage is relatively less prominent for fermented dough where more sheeting passes are necessary to produce a good quality product. Although no significant saving in energy has been observed during the development of fermented dough by sheeting, it has been found that the process is accompanied by an improvement in the bread crumb texture when compared to that of bread produced from conventionally mixed doughs (Stenvert *et al.*, 1979).

The efficiency of mixing achieved by sheeting over conventional mixing equipment can be highlighted by a hypothetical calculation applied to an individual layer thickness. For example, a 12.5 mm thick dough sheet passed through rollers with a 1.7 size reduction ratio, folding a half-sheet over itself and rotating it by 90° would yield more than a million layers with an average thickness for a single layer of approximately 0.6 μm. In terms of

size comparison a typical starch granule size is approximately 5 μm; hence the dough structure following extensive sheeting and 20 folds may lead to a two-dimensional cross-linked gluten network development with starch granules embedded in it. As shown in Figure 3.1, the number of sheeting steps applied to the dough sample has a significant impact on the volume and crumb structure of bread (Morgenstern *et al.*, 1999; Kilborn and Tipples, 1974).

It has been observed during the passing of the dough sheet through sheeting rolls that as the extent of reduction of the sheet thickness, referred to as 'reduction ratio', increases the swell and spread of the sheet also increases. However, the relationship between the dough physical characteristics and the reduction ratio becomes less predictive as the reduction ratio increases beyond 3.0 (Engman, Peck, and Wilson, 2005). Conversely, sheeting parameters (e.g. torque, the normal force between the two rollers, and the normal stress generated at the roller surface) measured on the rollers during the operation tend to decrease with an increase in the roll speed as a consequence of the shear-thinning rheological behaviour of dough. It has been demonstrated that most of these measured parameters can be related to the rheology of the dough as well as the physical characteristics of the sheet, notably its width (Reid *et al.*, 2001). Another important advantage of the sheeting operation is the redistribution of air bubbles, which appears, to impact the crumb quality (Stenvert *et al.*, 1979). The fine crumb structure obtained as a result of sheeting or rolling of the dough layer can be explained by the coalesce of entrapped air bubbles and their redistribution in the dough matrix, which later serve as nucleation centers for gas bubble formation during the baking stage. During sheeting as the dough is squeezed between two surfaces, the gas bubbles collapse and coalesce. Redistribution of air bubbles also take place during rolling due to the high pressure experienced by the dough under the roller surfaces, leading to partial dissolution of the bubbles and subsequent release of pressure, which leads to the generation of smaller size air bubbles. These small air bubbles improve the crumb structure uniformity as well as their fineness/quality.

Typical sheeting or shaping operations for plastic type or soft dough involves either depositing the dough in molds or shaping is attained during baking in ovens at high temperatures. Another approach is to use positive displacement equipment, which pushes the dough through a regular-shaped orifice and through an oscillating/moving cutting device (typically a metal wire); it cuts the dough and deposits the pieces into a baking tray. The third option is to use a roller with sharp edges on its surface to be able to cut sheets of dough into small individually shaped baked products (cookies); this technology is also referred to as a rotary molded cookie cutter.

Crackers or biscuits use a somewhat similar type of dough compared to cookie dough; however, their formulations include more fat and a lower moisture content. Due to the higher fat content there is a need for the formation of a gluten network that is able to hold the dough together during processing

as well as to entrap gas bubbles formed during baking. The cracker or biscuit dough has relatively more gluten development than cookies but it is lower than that in bread and buns.

Typical dough sheeting for the production of crackers and biscuits begins with dumping the dough from the mixer into the hopper. Here it is important to maintain the height of dough in the hopper, which exerts a certain pressure on the dough located at the bottom, resulting in uniform sheeting that especially avoids cracks or holes in the case of recycling dough scraps from down the line.

Depending on the motion of the dough sheet with respect to the roller motion and the conveyor belt, the dispensing of dough is done using two typical arrangements, which are named front or back discharge. The front discharge arrangement (Figure 3.2a) provides support to the dough as it is sheeted and transferred on to the conveyor belt. This system is helpful in handling short or less viscoelastic dough, which needs to be supported during its transfer from the sheeting rollers to the baking zone. The back discharge arrangement is illustrated in Figure 3.2b. It is useful for handling more viscoelastic dough, that is, a dough that can be stretched without damaging its smooth surface. Unlike the front discharge arrangement, the back discharge one includes a conveyor moving at a speed that is somewhat faster compared to the bottom roller. This is necessary to avoid a uniform transfer of the dough sheet to the conveyor band without creasing or crumpling. Conversely, due to the weak nature and less extensibility of doughs handled by front discharge arrangements, the conveyor speed runs at a slower speed than the bottom rollers in order to avoid crack and tear development. Alternate arrangments with two or four rollers are also available, but the three-roller arrangement is widely used.

The speed of the conveyor band carrying the dough towards and away from the sheeting rollers needs to be monitored precisely to avoid tearing or

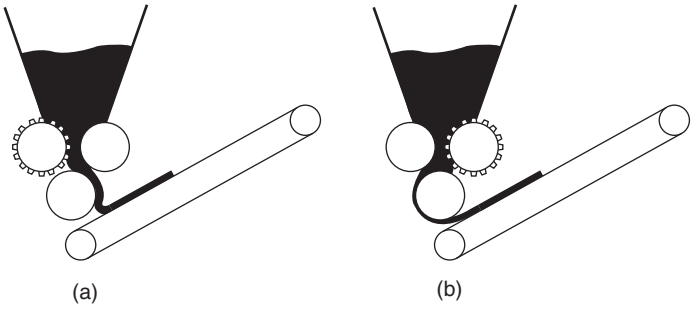

Figure 3.2 Three roller configuration delivery of dough from the sheeting equipment to the conveyor: (a) front discharge dough sheeting arrangement and (b) back discharge dough sheeting arrangement

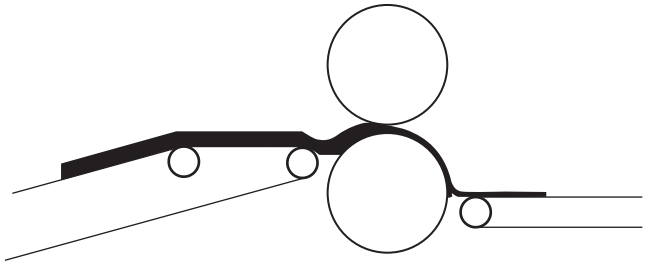

Figure 3.3 Position of feed and discharge conveyor band ends to dough sheeting rolls

crumpling of the dough sheet. It is advisable to allow some slack at the feed side as well as at the discharge side. This helps to avoid the development of internal stress or tear due to tension resulting of pulling effects created by the rollers at the feed point or by the conveyor band at the point of discharge (Figure 3.3). For large industrial systems, laser proximity sensors coupled with feeding and discharge conveyor band motor speed regulators can ensure continuous monitoring of the sheeting and discharge processes.

Lamination is an important industrial practice during cracker dough sheeting due to its multiple advantages. First, laminating (folding the dough sheet over itself) can repair cracks or tears developed during the sheeting operation. Second, by rotating the sheeting direction at a 90° angle after dough folding ensures uniform redistribution of stresses in two different directions. Further, repeated folding ensures uniform mixing of dough and development of the gluten network. Lamination also provides the opportunity of introducing another material (such as fat or sugar syrup) between dough layers to impart unique fluffy and layered characteristics to the final baked product. A manufacturing line of filled pastries can be devised using extrusion of dough and filling in to a tube, which then is flattened and manipulated to the desired shape using various configurations of the laminating line (for example, the line Rheon® for producing pastry dough used in meat pies).

It has been reported that changing the dough sheet orientation by 90° while passing between successive sets of sheeting rolls does not significantly affect the sheeting performance in terms of the resulting thickness, roll force, torque, or normal stress arising during the sheeting operation. However, the lateral spread of the dough sheet, which is measured as the percent increase in width, is observed to be negligible. This can be attributed to the viscoelastic nature of the dough, which tends to minimize stresses in the sheeting direction by expanding in a direction perpendicular to the sheeting rolls. These observations can be used to explain the use of folding back the dough sheet by artisan bakers as well as the use of 'cross-rollers' between a set of sheeting rolls in an industrial sheeting line to minimize the stress on the dough.

One significant difference between the sheeting and laminating operations is the presence of mixing as a result of the recirculation flow produced in the feed section of the rollers. Some degree of mixing is desired during the sheeting whereas it is considered detrimental during lamination.

Morgenstern and Ross (1998) found that when the dough sheet is passed through a gap that is larger than a fourth of the sheet initial thickness, there is a negligible recirculation flow. However, as the gap between the rollers decreases a recirculation flow is always observed. As a rule of thumb, when the gap between the rollers is smaller than an eighth of the original dough sheet thickness, flow recirculation becomes evident (Figure 3.4).

This behavior is not unique to doughs. Pearson (1985) and Middleman (1977) reported similar observations for polymer materials caused by recirculation flows generated near the region of contact between the feed sheet of the viscoelastic material and the roller.

In addition to the backflow mixing observed in the feeding of the rollers there are other effects produced by the viscoelasticity of the dough that affect the thickness of the final sheet. For instance, an increase in the reduction ratio, the ratio between the gap among rollers and the dough thickness, results in an increase in the sheet thickness due to the snack back effect produced by the viscoelastic dough after passing the rollers (Ren, Walker, and Faubion, 2008). One of the common drawbacks encountered during high size reduction in single pass sheeting is likely to be produced by the viscoelasticity of the dough, which generates the rippled surface of the rolled sheet. In a series of articles describing the flaking process, a process having some similar features to sheeting, Levine *et al.* (2004, 2003, 2002a, 2002b) demonstrated the changes in the properties of the final product as a consequence of the dough viscoelasticity.

Scale-up of sheeting operations is an important task in the design of new processes and products. Levine (1985) used an empirical approach to describe the sheeting operation and applied a dimensional analysis to the experimental data to generate a mathematical model that relates the final dough sheet

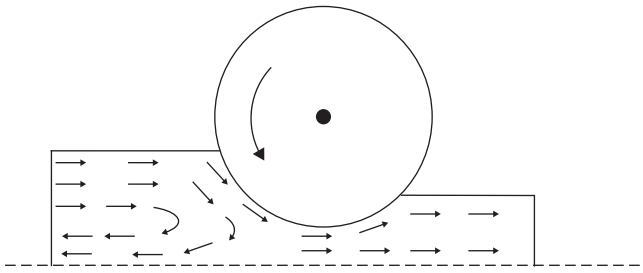

Figure 3.4 Schematic illustrating the recirculation of dough generated at the nip of the rollers when the dough sheet enters between the rollers for conditions of small gaps between the rollers (source: Love 2003, p. 4. Reproduced with Permission of R. Love)

thickness to the roll gap and roll diameter:

$$\frac{t_0}{t_g} \cong 0.06\left(\frac{t_g}{D}\right)^{-0.58} + 1 \tag{3.1}$$

Here t_0 is the final dough sheet thickness, t_g is the gap between rollers, and D is the diameter of the rollers. Calculations based on this model are reported in Table 3.1, where the final dough thickness was calculated by assuming an initial thickness of the sheet of 1/8. Based on these calculations, it is possible to estimate the percentage of spring-back as the increase in dough sheet thickness from the sheeting operation expressed as a percentage value of the roller gap. The table clearly illustrates that as the sheeting roll diameter increases, the percentage of spring-back also increases. This is supported by the fact that as the sheeting roll diameter increases the amount of energy input also increases, which in turn induces larger residual stresses in the dough promoting spring-back. As the dough sheet comes out of the sheeting roll, the stresses developed in the dough sheet are relaxed and can be measured in terms of spring-back. Based on the percentage of spring-back values reported in Table 3.1, it can be safely concluded that the use of sheeting rolls with smaller diameters produces doughs with smaller residual stresses, which is likely to require less residence time. The practical advantage of having doughs with these characteristics is that shorter conveyor belts are required between two sheeting stations as well as the length of the conveyor belt going to the baking oven.

The percentage spring-back does not change significantly with the strain rate, as illustrated in Figure 3.5. Sheeting operations for viscoelastic dough differs from plastic dough because the elastic component of the viscoelastic dough promotes the recovery of the shape once the force is removed. Thus, recovery of the material after the applied strain or stress needs to be considered when designing the process line such that the desired final product shape is achieved. This tendency of regaining the shape makes the handling of the viscoelastic dough more difficult to process compared to a plastic-like dough where a simple wire cutter, pressing on to the mold, or depositing in

Table 3.1 Spring-back percentage calculated from data generated by the simulation model (source: Levine 1985. Reproduced with permission of John Wiley & Sons, Ltd)

Diameter (inch)	Roller gap (inch)	Final dough sheet thickness	Spring-back (%)
3	0.085	0.125	47.1
4	0.079	0.125	58.2
6	0.070	0.125	78.6
8	0.062	0.125	101.6
12	0.052	0.125	140.4

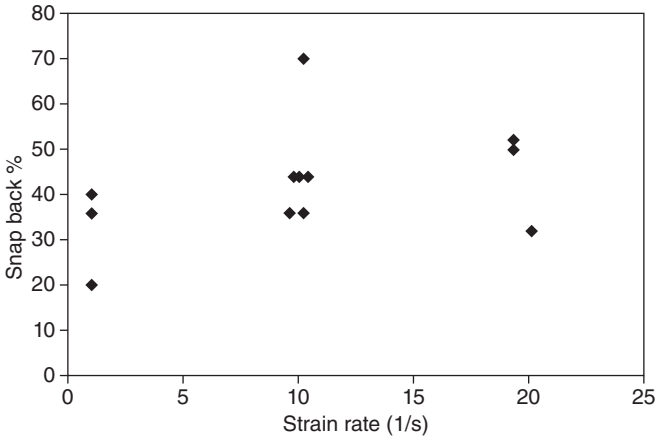

Figure 3.5 Percentage of snap-back of the dough sheet after passing through the sheeting roll as a function of the strain rate at the sheeting roll (source: Adapted from Kempf 2005. Reproduced with permission of AACC International)

the mold serves the purpose of portioning the dough into an individual item before baking.

In addition, for viscoelastic doughs one of the major considerations is releasing the internal stresses generated during shaping/sheeting before it goes for baking. This is important to avoid losing the shape of the dough piece. As the dough temperature rises during the initial stages of baking, the consistency of the dough is reduced and it could easily happen that any residual stresses could alter the shape of the dough piece before crumb development and setting of the structure. This is the reason when handling viscoelastic doughs for using longer conveyor belts and allowing time for relaxation of internal stresses developed during sheeting/shaping before the individual product enters the baking oven. An alternate approach to improve the shape retention is to use sheeting/shaping operations that do not result in high internal stress development. For example, some companies advertise lines with zero stress applied to the dough.

Other recent developments in sheeting equipment available in the market rely on this principle where small deformations are applied to dough over numerous steps rather than large deformation and fewer passes between the sheeting rollers. One example of the gentle sheeting dough handling process is the stress-free rolling process offered by RONDO. The patented 'Smartline' system by RONDO (Figure 3.6) applies multiple small rollers over small distances, which avoids larger strains and larger stresses developing in the dough mass as a result. Thus, the dough is spread/sheeted with multiple smaller rollers working over small distances. To achieve stress-free stretching of the dough sheet, a set of smaller diameter rollers at a suitable angle comes

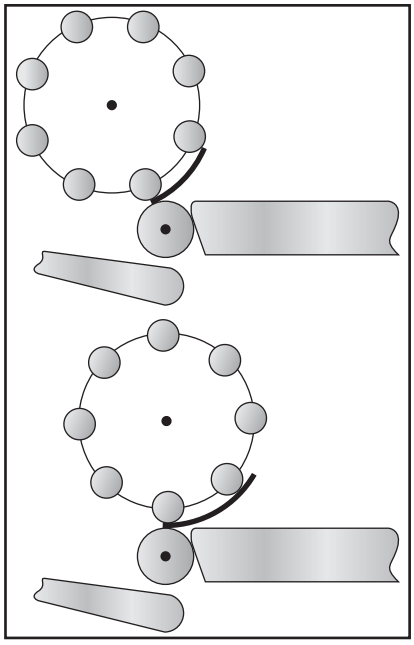

Figure 3.6 Schematic drawing for the 'Smartline' sheeting system from RONDO (source: Reproduced with permission of RONDO)

in contact with the top dough surface, while the bottom part moves over a relatively larger roller. The speed of the top rollers and bottom roller can be set depending on the type of dough handled by the system. The resulting sheet of dough has low residual stress development due to gentle stretching of the dough. This approach fits well with the typical rheological behavior of the dough.

During sheeting the dough sheet undergoes deformation with respect to the height, which is easily visible as a change in dough sheet thickness. However, changes in length and width are difficult to visualize, especially in continuous operations at the commercial level. Love (2003) showed experimentally how these changes in width and length become manifest in a dough sheet during sheeting (Figure 3.7). This visualization promotes the need to gain an understanding of the origin of undesirable changes due to deformation during the sheeting operation.

Since pressure and shear forces have been associated with de-aeration and quality of dough products it can be safely assumed that the properties of the sheeted material may differ between the center and the edge. Levine *et al.* (2002a) proposed a numerical approach to simulate dough behavior during sheeting, similar to the calendering of finite width sheets between two rollers. Simulating the spread of the dough sheet as a function of the ratio W_0/H_0

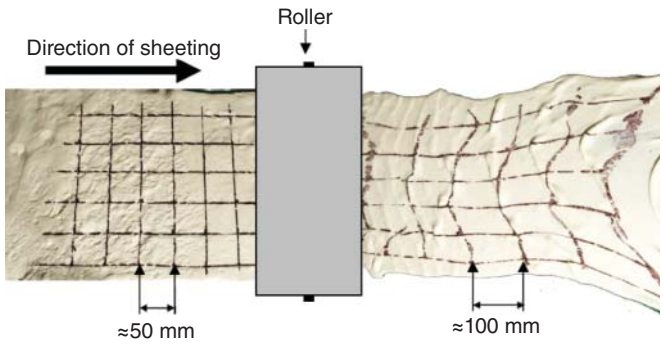

Figure 3.7 Deformation in the dough sheet during the sheeting operation showing damage to the dough surface as well as changes in the shape of the sheet (source: Love 2003, p. 3. Reproduced with Permission of R. Love)

(W_0 and H_0 are the initial width and the thickness of the sheet, respectively) showed that the edges of the sheet exhibit high pressure gradient regions. The presence of high gradients would stretch the material near the edge leading to dough sheet tearing. This type of tearing can be reduced by laminating the dough in a typical cross-crossing direction, as mentioned above.

3.2.2 Rheological studies on dough behaviour

Dough rheology helps to determine various parameters relating the viscoelastic behavior of dough, such as storage and loss moduli, and viscous parameters, such as flow consistency and the flow behavior index, with characteristics of the dough process (defined below). Two types of tests have been used to characterize doughs, which are classified in terms of the magnitude of the applied strain or stresses as small and large deformation tests. Small strains/stresses in small deformation tests are generally applied to find interactions among the components of the dough, notably the moisture content and type of flour, whereas large deformation tests are used to characterize the properties of dough and how they interact with the process (Kim *et al.*, 2008; Zheng *et al.*, 2000). Thus, rheological studies provide tools to characterize various types of dough as well as ability to simulate the performance of dough under various processing conditions or for developing quality control methods (Dobraszczyk and Morgenstern, 2003).

Most food materials are viscoelastic and therefore their properties depend on the time under which the stress or the strain is applied to the material. This is important in many aspects of dough processing; for instance, if the dough is deformed quickly, such as in mixing or sheeting, then the rheological properties of dough will be very different in magnitude than those observed when the dough material is deformed or stressed continuing to produce any sort

of steady flow. The inherent viscoelasticity of doughs adds complexity to the design and optimization of dough processing because, depending on the magnitude of the deformation and the deformation rate, the rheological properties of the dough may impact differently on the process. This is important especially when using values measured at lower strain rates to model the behavior of the dough at higher strain rate operations, which can lead to an increase of up to a couple of orders of magnitude. That has been observed when no traditional mechanically and shear-based techniques are used to characterize the dough material, especially ultrasound techniques that are characterized by significantly higher frequencies and type of deformation (Campanella, 2010; Leroy *et al.*, 2010; Mert and Campanella, 2008; Lee, Pyrak-Nolte, and Campanella, 2004; Ross, Pyrak-Nolte, and Campanella, 2004; Létang *et al.*, 2001). Other novel rheological methods, such as the impulse technique, are related to ultrasonic measurements of dough properties during processes in which the mechanical properties of the dough can be significantly affected by the extent of the deformation, notably fermentation (Lee and Campanella, 2013). Although ultrasound frequencies are much higher than those used in mechanical rheometry, the ultrasonic characterization of doughs has provided relevant and key data on dough properties. It also offers the potential to be used as an online rheological technique to characterize the material during processing.

The other aspect to be taken into account in dough rheology is the type of deformation under which the dough sample is tested. During processing, dough is subjected to either shear (e.g., in a mixer) or extensional strains (sheeting, fermentation) or a combination of both deformations. Thus, if a relationship between rheological properties and their impact on processing is sought, it becomes necessary to use rheological tests that use these types of deformations. Zheng *et al.* (2000) reviewed and applied a variety of rheological tests, including shear and extensional deformations, to monitor changes of dough during dough development.

As discussed above, another important area in dough rheological testing is the large deformation test. Large deformation tests employ relatively higher strain levels and explore the rheological behavior of dough in the nonlinear viscoelastic region. The data generated from large deformation tests are of practical importance, that is, it can explain and model the dough behavior under commercial processing conditions encountered by dough on baking lines. These tests help to characterize the dough for various baked goods objectively rather than by using subjective descriptors, such as soft and hard dough for breads and biscuits or nonsticky and sticky dough for crackers and cookies. These tests can help to incorporate science in addition to the baking process to elevate it from just being a form of art and at the same time helping to open the possibility for other applications, including the design of the baking system such as sheeting, rolling, and flattening (Bhattacharya, 2010; Castell-Perez and Steffe, 1992). The potential of better process control is also closely associated with a correct characterization of the dough material. Large

deformation tests as well as recoil and relaxation behavior tests of dough can help to describe the rheological behavior of the dough material when it moves from the mixer to the baking oven.

Many researchers have attempted to simulate the dough flow behavior using a number of rheological models. Early attempts were made using the Newtonian model, which was found to be inadequate to account for the nonlinearity in the flow behavior of dough. One of the most widely used rheological models is the power law model given by the following equation:

$$\eta = k\dot{\gamma}^{n-1} \tag{3.2}$$

where η is the apparent viscosity, $\dot{\gamma}$ is the shear rate, k is the consistency index, and n is the flow behavior index. The consistency index has a unit of Pa s^n whereas the flow index is unitless. Values of consistency and flow indexes reported range from 2000 to 50 000 Pa s^n and 0.25 to 0.5, respectively. They depend on the dough formulation including moisture content, the type of flour, and the presence of a plasticizer. Although this variation can be the result of different formulations it clearly demonstrates the wide range of behaviour exhibited by dough and the impact these properties may have on dough processing. The role of water as a plasticizer in dough is critical in modeling dough behavior (Faubion and Hoseney, 1990). It is also possible to develop a rheological model to accommodate specific formulation attributes such as salt and fat content (Raghavan *et al.*, 1995). Launay (1990) reported the use of the Kelvin-Voigt model (a dashpot-liquid element representing liquid behavior in parallel with a spring representing elastic behavior) for describing the nonlinear and power law behavior of dough. However, the Kelvin-Voigt model is a solid viscoelastic model and dough has properties that resembles more the properties of a liquid. Thus, the most widely used technique to model dough rheological behavior is the Maxwell (Bagley, Christianson, and Martindale, 1988) or generalized Maxwell model (Wang and Kokini, 1995). These approaches involve the use of Hookean (spring) and Newtonian (dashpot) elements in series configurations to accurately represent the observed rheological behavior of dough. The models provide information on the behavior of dough under a small deformation but fail to describe the behavior of dough at large deformations. Other models, like, for instance, models such as those described by Wang and Kokini (1995), and more recently by Carriere, Thomas, and Inglett (2002) and Lefebvre (2006), are necessary to describe the behavior of dough under nonlinear viscoelastic conditions. Regardless of the rheological method, the understanding of dough rheological behavior under large deformations can be very useful in explaining the typical behavior of dough under processing. Amemiya and Menjivar (1992) analyzed the response of dough by comparing the behavior with that of filled elastomeric systems. The rheological response was

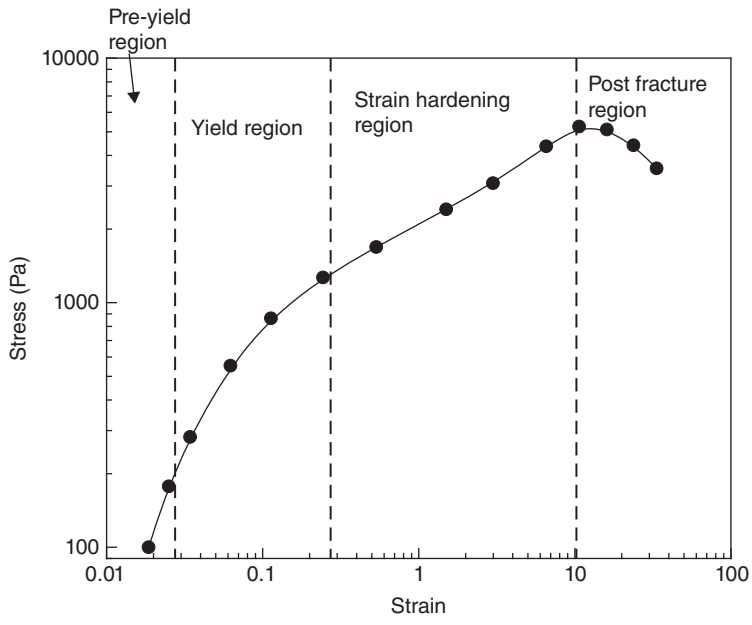

Figure 3.8 Typical characterization of a dough material (source: Amemiya and Menjivar 1992. Reproduced with permission of Elsevier)

divided into four distinct regions named pre-yield, yield, strain hardening, and post-fracture (Figure 3.8). The pre-yield behavior is observed over very small strain levels below 3.0% strain where starch-starch and starch-protein interactions dominate the dough response to a shear deformation. Beyond the pre-yield region, as the strain increases, the dough exhibits typical yield behavior, that is, a decline in the stress-strain curve slope is observed. The yield region of the stress-strain curve can be explained by dough overcoming the short-range interactions as larger deformation is experienced. The yield region is followed by a strain hardening region where the level of stress starts to increase with strain typically observed between 0.25 to 13. With the increase in strain above 0.25, the short-range interactive forces are overcome and plastic flow is initiated. However, other long-range forces may come into play at this strain level (i.e., the deformation is moving from microscopic or micrometer displacement to macroscopic or larger than a millimeter displacement) that are the result of the well-developed protein network that exists at the macroscopic level. These long-range protein-protein interactions appear to give rise to a typical strain hardening effect encountered during processing of the dough, where the gluten network is well developed during dough mixing, for example, for breads. Conversely, for baked goods where gluten development is not desired, for example biscuits and cookies, the dough often does not exhibit the strain hardening effect since the biscuit dough does not have

large-scale protein-protein interaction due to a poorly developed gluten network.

Finally, as the strain increases beyond ~13 the stress reaches a peak and decreases, which can be due to breaking of the protein network. Typically at such high strain levels the dough starts to break, which is not desired, especially for any operations involving rolling, sheeting, shaping, or flattening of the dough. These tests have been used to characterize the behavior of different types of doughs, like, for example, those used in the manufacture of breads and biscuits (Figure 3.9). In this test the viscosity of the dough sample was measured at a low shear rate of 1/s and the evolution of the shear and normal stresses were followed over time. Values of normal stresses measured on the rotating plate (in parallel plate geometries) or the rotating cone (in cone and plate geometries) are indicative of the dough elasticity; the largest is the normal stress, the largest is the elasticity of the sample. As illustrated in Figure 3.9, the shear stress evolution follows the general trend described in Figure 3.8. It also illustrates that bread dough is more elastic than biscuit dough.

Xiao, Charalambides, and Williams (2007) were able to generate numerical models to follow the dough behavior during sheeting at relatively low reduction ratios (lower than 1.7). They generated a numerical model similar to the one proposed by Goh, Charalambides, and Williams (2005, 2003) using the finite element numerical method and using material and rheological

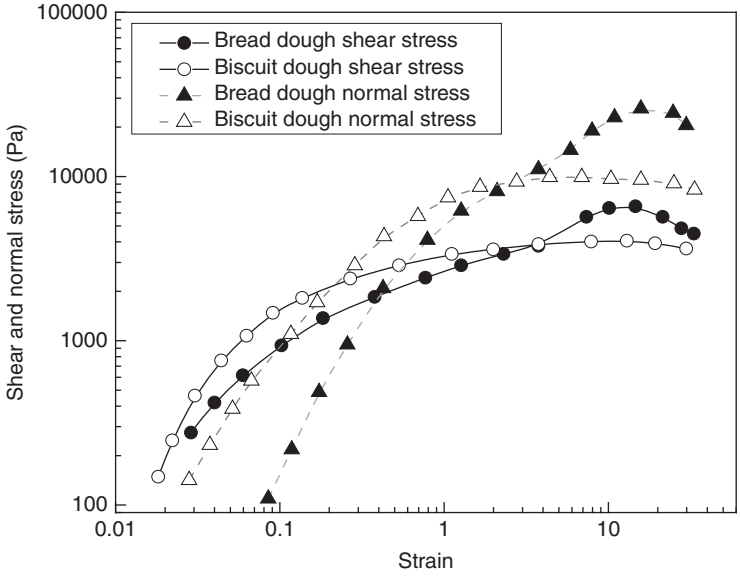

Figure 3.9 Different types of dough behaviors as a function of strain level (source: Amemiya and Menjivar 1992. Reproduced with permission of Elsevier)

parameters gathered from various rheological tests, such as uniaxial compression, the loading-unloading test, and the stress relaxation test. The model was found to predict successfully the dough behavior during the two-step rolling processes provided the volume flow rates between the steps of rolling were the same. The difference in the volume flow rate between two rolling stations led to the generation of significant amounts of compressive or tensile stresses between the rolling stations, which were not included in the numerical model.

3.3 Shaping

Typical shaping operations for less viscoelastic doughs (e.g., cookie, shortbread, crackers, etc.) consist of the molding of the sample. The dough is either forced into the mold to achieve the desired shape or sheeted in a desired thickness and then shaped by cutting with a rotary or pneumatic mold cutter. Mechanical devices can also be used to punch out the final shape, which is the case for cookies, crackers, and biscuit processing.

Depending on the type of dough, the shaped pieces can be removed pneumatically from the conveyor band or the baking sheet by using either pressurized air to blow out the pieces on to another conveyor belt or by using a partial vacuum so that the individual pieces can be lifted and transferred to the next conveyor band. Once the shaped dough is transferred to the baking tray or conveyor band to the baking oven, the excess dough left behind is collected as scrap and mixed with new dough before the sheeting operation. When using recycled dough, it is important to keep in mind that the scrap dough generated has already been processed. Thus, it is important to estimate properly how re-mixing it with new dough will influence the dough behavior during processing.

Regarding the cutting of the dough piece to achieve the desired shape, it has been demonstrated that ultrasonic vibrations of the cutting blade help to obtain clean cuts of dough, which is very helpful, especially when dealing with sticky doughs. Ultrasonic knives vibrating up and down about 20 000 times per second are able to slice a product as it comes out of an oven or down a conveyor, or slit the product into bar-shaped portions. This high-frequency vibration creates a nearly friction-free surface between the tool (e.g., knife, diverter fence, or transfer bar) and the product, so problems with sticking and pinching simply disappear. With the greatly reduced friction, there is virtually no knife abrasion. Tools stay sharper longer, reducing annual maintenance costs. It also increases productivity by minimizing the production time previously lost during cleaning the machinery. It has been also demonstrated that this technology can totally eliminate procedures strictly necessary when using conventional cutting technology, such as pre-cooling or heating the product before the cutting operations, or realigning the product after cutting prior to the packaging operation.

For a typical viscoelastic dough (e.g., breads and buns), as discussed earlier, the residual stress arising from the dough elasticity becomes a critical factor in retaining the shape imparted, so a period of stress relaxation may be necessary prior to cutting. Another important factor to consider is that the dough is well mixed due to work input imposed by the mixing operation as well as during sheeting. Hence, work input during the shaping operation needs to be minimized to avoid undesirable effects of overmixing and weakening the gluten network, in particular when the dough has been fermented. A typical example is just before the baking operation when dough pieces are portioned as individual pieces of the desired weight/volume and the next step gives the piece the desired shape of the product.

Rounding is one form of shaping operation where a portion of dough having an irregular rough surface with a well-developed gluten network must be transformed into a regular desired shape for subsequent baking steps. The purpose of the rounder is to provide a smooth and dry-appearing thick skin around the pieces; it closes the open gas cells on the cut surface and stops gas diffusing out of the dough. Depending on the size and shape of the product bowl, umbrella or drum rounders are used in the baking industry. In the bowl-shaped rounder the dough piece is rounded by sliding it up along a spiral path or rail around the inside of a rotating bowl. The rounded piece finally emerges at the top where it has a smooth skin and a round shape. Another important function of the rounder is to align the randomly organized gluten network in the well-mixed dough in a regular pattern, which later emerges as an appealing grain structure in the finished product crumb.

For the umbrella-type rounder the bowl is inverted and the dough piece travels up the outer surface of the cone and enters at the base; the rounded dough piece exits at the apex. These two types of rounders are widely used in the baking industry; however, there are various other types of rounder less popular or used for specific product applications, for example, the drum style, where the conical bowl surface is replaced with a drum. Rounders also exist with concave sides where the dough piece is moved through multiple passes inside the cavity, resulting in a piece with a round shape. Typical examples of a product-specific rounder is the drum-type rounder for round or long buns. Here the dough is placed in a cavity on the outer drum and the concentric inner drum forms the bottom surface of the cavity, which rotates and gives shape to the dough portioned in the cavity. To provide grip in the dough, the inner drum surface can have rough surfaces or small flutes/corrugations along the length of the shape.

For viscoelastic dough with a dominant elastic behavior (e.g., tortilla, flat bread, naan, etc.) imparting a final desired shape is a very different process compared to that of typical bread or cookie shaping operations. The most common practice is to portion the dough followed by shaping and flattening before the baking or frying operation. The typical portioning operation involves the use of a combination of the metering auger with a reciprocating wire cutter.

The most common practice to control the portion size is to control the speed of the reciprocating arm that portions the dough. It is possible to increase the auger speed by pushing the dough through the cutter; however, due to the viscoelastic nature of the dough an uneven pressure buildup can be created due to these changes in the auger speed, which makes it difficult to control the portion size. Instead, changing the cutting mechanism speed is a better option. At this stage the portioned dough piece will have roughly a spherical shape, with two relatively flat ends where the cutting element separates it from the bulk of the dough. To achieve a spherical shape, the dough pieces are transferred to a conveyor where a top surface moving in circular motion comes into contact with the dough pieces and finishes the spherical shape (Figure 3.10).

An alternate arrangement of relatively solid doughs and semi-continuous operations can be achieved by using two plates, one of which is moving in circular motion while the other plate remains stationary. To shape more than one dough piece, a number of compartments can be designed to contain individual dough pieces. In this case the dough sheet, thick enough to give the individual piece of dough a desired weight, is loaded on to the bottom plate and is divided into individual pieces by a spring-loaded compartment pattern that serves a dual purpose that includes cutting. Then one of the plates compresses the dough piece a little further to apply a slight pressure and starts a circular motion to make the spherical dough ball mimick the motion of manually rolling the dough ball between two hand palms.

The *flattening operation* typically follows the dough-shaping operation where the dough piece is rounded into spherical or cylindrical shapes, which is necessary to get the circular flattened dough. This operation is typically for tortilla or flat bread making where the elastic behavior dominates the viscous behavior. Consideration of the dough spring-back is also important at this stage, which is the same as during sheeting. To maintain the uniformity on the dimension for individual product portions, both the extent of pressure applied and the time duration of the pressing are important parameters. Here, it is possible to use insights gained from the dough rheology to determine both the pressure and the time of the flattening operation. It is important to know the rate of stress relaxation and strain recovery of the material, which

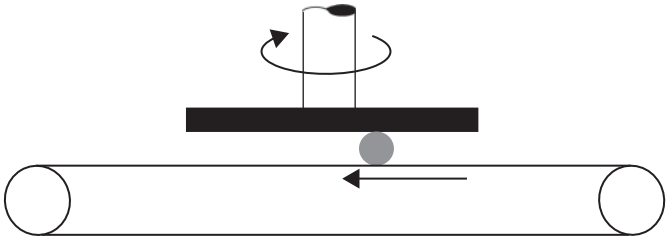

Figure 3.10 A conveyor band and a top plate for rolling dough into a spherical ball

can be obtained from experiments that resemble the operation. To allow the dissipation of internal stresses, the flattening can be achieved in two steps with some relaxation time in between. This two-step flatting gives better shape control compared to the single-step flattening process.

Flattening of softer dough with plastic/viscous behavior is very difficult using the above described setup. Interestingly there are few patents and commercial devices available to flatten soft doughs. One typical example is pizza dough, which is very delicate due to extensive fermentation during the dough making, which is very critical in flavor development. To flatten the piece of dough coming from either the divider or the rounder, it is positioned in a flattening station below two or more than two conical rollers positioned with the apex in the center and free to rotate. The typical flattening station operation involves lowering the cone assembly slowly on the piece of dough while it is rotating in a circular motion. Each roller is rotating while moving in a circular motion and gradually compresses the dough piece to the final shape of the circular flattened pizza dough. One such process line from Rheon Automatic Machinery GmbH can handle pizza dough from 5 to 12 inches in diameter in various shapes, like round, rimmed, or oval. Further, specialty pizzas like calzone or Stromboli can also be stretched on this pizza spinner with additional accessories. The Stress Free® pizza dough stretcher machine from Rheon can shape dough portions into a pizza base. The portioned dough from either the divider or rounder is transferred to the conveyor where the dough is pressed against two or more cones rotating with the apex at the center. These cones virtually duplicate the hand-tossing technique to create high-quality pizza crusts with even thickness. The rotation can be either in a single direction, that is, clockwise/counterclockwise, or in both directions. This equipment can shape pizzas in round, rimmed, and oval shapes from 5 to 12 inches in size, as well as specialty pizzas like calzone or Stromboli.

References

Amemiya, J.I. and Menjivar, J.A. (1992) Comparison of small and large deformation measurements to characterize the rheology of wheat-flour doughs. *Journal of Food Engineering*, **16**(1–2), 91–108.

Bagley, E.B., Christianson, D.D., and Martindale, J.A. (1988) Uniaxial compression of a hard wheat-flour dough – data-analysis using the upper convected Maxwell model. *Journal of Texture Studies*, **19**(3), 289–305.

Bhattacharya, S. (2010) Stress relaxation behaviour of moth bean flour dough: product characteristics and suitability of model. *Journal of Food Engineering*, **97**(4), 539–546.

Campanella, O.H. (2010) Instrumental techniques for measurement of textural and rheological properties of foods. In: *Emerging Technologies for Evaluating Food Quality and Food Safety* (Cho, Y.J. and Sukwon Kang, S., eds). CRC Publisher, pp. 6–53.

Carriere, C.J., Thomas, A.J. and Inglett, G.E. (2002) Prediction of the nonlinear transient and oscillatory rheological behavior of flour suspensions using a strain-separable integral constitutive equation. *Carbohydrate Polymers*, **47**, 219–231.

Castell-Perez, M.E. and Steffe, J.F. (1992) Using mixing to evaluate rheological properties. In: *Viscoelastic Properties of Foods* (Rao, M.A. and Steffe, J.F., eds). Elsevier Applied Science Publishers, Barking, England, pp. 247–284.

Dobraszczyk, B.J. and Morgenstern, M.P. (2003) Rheology and the breadmaking process. *Journal of Cereal Science*, **38**(3), 229–245.

Engman, J., Peck, M.C. and Wilson, D.I. (2005) An experimental and theoretical investigation of bread dough sheeting. *Food and Bioproducts Processing*, **83**(3), 175–184.

Faubion, J. M. and Hoseney, R. C. (1990) The viscoelastic properties of wheat flour doughs. In: *Dough Rheology and Baked Product Texture* (Faridi, H. and Faubion, J., eds). Springer, USA, pp. 29–66.

Goh, S.M., Charalambides, M.N. and Williams, J.G. (2003) Mechanical properties and sensory texture assessment of cheeses. *Journal of Texture Studies*, **34**(2), 181–201.

Goh, S.M., Charalambides, M.N. and Williams, J.G. (2005) Characterization of the nonlinear viscoelastic constitutive properties of mild cheddar cheese from indentation tests. *Journal of Texture Studies*, **36**(5–6), 459–477.

Kempf, T.P., Labuza, T.P. and Levine, L. (2005) Elastic recovery of a wheat flour dough during sheeting. *Cereal Foods World*, **50**(6), 316–320.

Kilborn, R.H. and Tipples, K.H. (1974) Implications of the mechanical development of bread dough by means of sheeting rolls. *Cereal Chemistry*, **51–55**, 648–657.

Kim, Y.-R., Cornillon, P., Campanella, O.H., Stroshine, R.L., Lee, S. and Shim, J.-Y. (2008) Small and large deformation rheology for hard wheat flour dough as influenced by mixing and resting. *Journal of Food Science*, **73**(1), E1–E8.

Launay, B. (1990) A simplified nonlinear model for describing the viscoelastic properties of wheat-flour doughs at high shear strain. *Cereal Chemistry*, **67**(1), 25–31.

Lee, S.Y. and Campanella, O. (2013) Impulse viscoelastic characterization of wheat flour dough during fermentation. *Journal of Food Engineering*, **118**(3), 266–270.

Lee, S.Y., Pyrak-Nolte, L.J. and Campanella, O.H. (2004) Determination of ultrasonic-based rheological properties of dough during fermentation. *Journal of Texture Studies*, **35**(1), 33–52.

Lefebvre J. (2006) An outline of the non-linear viscoelastic behavior of wheat flour dough in shear. *Rheologica Acta*, **45**, 525–538.

Leroy, V., Pitura, K.M., Scanlon, M.G. and Page, J.H. (2010) The complex shear modulus of dough over a wide frequency range. *Journal of Non-Newtonian Fluid Mechanics*, **165**(9–10), 475–478.

Létang, C., Piau, M., Verdier, C. and Lefebvre, L. (2001) Characterization of wheat-flour-water doughs: a new method using ultrasound, *Ultrasonics*, **39**(2), 133–141.

Levine, L. (1985) Throughput and power consumption of dough sheeting rolls. *Journal of Food Process Engineering*, **7**, 223–238.

Levine, L. (1996) A model for the sheeting of dough between rolls operating at different speeds. *Cereal Foods World*, **41**(8), 690–697.

Levine, L., Corvalan, C.M., Campanella, O.H. and Okos, M.R. (2002a) A model describing the two-dimensional calendering of finite width sheets. *Chemical Engineering Science*, **57**, 643–650.

Levine, L., Campanella, O.H., Corvalan, C.M. and Okos, M.R. (2002b) A model for the formation of multiple flakes during cereal flaking. *Cereal Food World*, **47**, 210–223.

Levine, L., Campanella, O.H., Corvalan, C.M., Okos, M.R. and Gonzalez, D. (2003) A model for predicting the aspect ratio of cereal flakes. *Cereal Foods World*, **48**, 289–299.

Levine, L., Campanella, O.H., Okos, M.R. and Ross, K. (2004) Observations on physicochemical changes with flaking. *Cereal Food World*, **49**, 65–70.

Love, R.J. (2003) Mathematical modelling of bread dough sheeting. PhD Thesis, Massey University, Palmerston North, New Zealand.

Mert, B. and Campanella, O.H. (2008) The study of the mechanical impedance of foods and biomaterials to characterize their linear viscoelastic behavior at high frequencies. *Rheologica Acta*, **47**, 727–737.

Middleman, S. (1977) *Fundamentals of Polymer Processing*. McGraw-Hill Ltd, New York, USA.

Morgenstern, M. and Ross, M. (1998) Sheeting of dough. Research Bulletin, Crop and Food 3, Winter.

Morgenstern, M.P., Zheng, H., Ross, M. and Campanella, O.H. (1999) Rheological properties of sheeted wheat flour dough measured with large deformations. *International Journal of Food Properties*, **2**(3), 265–275.

Moss, H.J. (1980) Strength requirements of doughs destined for repeated sheeting compared with those of normal doughs. *Cereal Chemistry*, **57**(3), 195–197.

Pearson, J.R.A. (1985) *Mechanics of Polymer Processing*. Elsevier Applied Science Publishers, London, England.

Raghavan, C.V., Chand, N., Srichandan Babu, R. and Srinivasa Rao, P.N. (1995) Modeling the power consumption during sheeting of doughs for traditional Indian foods. *Journal of Food Process Engineering*, **18**(4), 397–415.

Reid, J.D., Corvalan, C.M., Levine, L., Campanella, O.H. and Okos, M.R. (2001) Estimation of final sheet width and the forces and power exerted by sheeting rolls. *Cereal Foods World*, **46**(2), 63–69.

Ren, D.Q., Walker, C.E. and Faubion, J.M. (2008) Correlating dough elastic recovery during sheeting with flour analyses and rheological properties. *Journal of the Science of Food and Agriculture*, **88**, 2581–2588.

Ross, K.A., Pyrak-Nolte, L.J. and Campanella, O.H. (2004) The use of ultrasound to characterize the effect of mixing time on the rheological properties of dough. *Food Research International*, **37**(6), 567–577.

Stenvert, N.L., Moss, R., Pointing, G., Worthington, G. and Bond, E.E. (1979) Bread production by dough rollers. *Bakers Digest*, April, 22–27.

Wang, C.E. and Kokini, J.L. (1995) Prediction of the nonlinear viscoelastic properties of gluten doughs. *Journal of Food Engineering*, **25**(3), 297–309.

Xiao, W., Charalambides, M.N., and Williams, J.G. (2007) Sheeting of wheat flour dough. *International Journal of Food Science and Technology*, **42**(6), 699–707.

Zheng, H., Morgenstern, M.P., Campanella, O.H. and Larsen, N.G. (2000) Rheological properties of dough during mechanical dough development. *Journal of Cereal Science*, **32**(3), 293–306.

4

Extrusion Processing of Foods

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4.1 Introduction

An extruder is a machine that houses an Archimedes screw that rotates in a tightly fitting stationery barrel. Several applications in metal and polymer processing systems have encouraged the food professionals to look into this technology carefully and apply for processing food materials. The slow start in food extrusion technology in the 1970s has flourished and now has applications in various processing systems and commercial products. Application of the extrusion system for forming pasta products by manual means is more than 100 years old technique. However, heating the raw materials during extrusion opens the possibility of manufacturing several ready-to-eat (RTE) or ready-to-cook (RTC) foods. The heating of the product can be accomplished in several ways. These are the dissipation of mechanical energy by high-speed rotation of the screw inside the tightly fitting barrel and thermal input through steam injection and by using heating systems mounted on the external surface of extruder barrels. In practical terms, the present day extruders work on the principle of thermomechanical and external thermal inputs that cook the moist raw material and thus the name 'extrusion cooking' is widely used by technologists and applied efficiently for the manufacture of several food products on a commercial scale.

The advantages of extrusion cooking are the high production capacity, performing a number of unit operations together, energy efficiency, processing of

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relatively dry ingredients of foods, reduced manpower requirement and floor space, and a continuous processing facility – all these factors decrease the cost of production to make the processing system economically pragmatic. Further, as extrusion technology operates with the principle of a high-temperature short-time (HTST) processing system, the nutritional damage is minimized while the inactivation of microbes and undesirable enzymes are maximum to make the product safer.

Several books are available that deal with food extrusion and extrusion cooking of foods. The contributors are Maskan and Altan (2011), Guy (2001), Riaz (2000), Chang and Wang (1998), Steven and Covas (1995), Frame (1993), Benbow and Bridgwater (1993), Kokini, Ho and Karwe (1992), Mercier, Linko and Harper (1989), Jowitt (1984), Harper (1981a, 1981b) and Janssen (1978, 2004). Further, a number of review articles are available concerning specific aspects of food extrusion. These describe nutritional aspects of extruded foods (Camire, 2000), quality aspects of extruded foods (Riaz, 2000), the raw material used for extrusion (Bhattacharya, 2011), newer applications of extrusion (Jorge, Alvarez and Rao, 2011), mathematical treatments (Barron *et al.*, 2002) and structural features of the products (Moscicki and Wojtowicz, 2011).

A number of nutritional intervention programmes are linked with the application of extrusion technology wherein the overall target is to provide low-cost nutritious high-protein cooked foods. The corn-soy-skim milk (CSM) is just one example of the product to offer a balanced food for people with nutritional deficiencies (Harper, 1981b). In earlier days, an extruder cooker has been a costly machine but at present it is manufactured in several countries and the price has reduced drastically. However, with continuous improvement in the design of an extruder and the development of new products, this technology is still young and it is early to feel its presence, but it is competing well with other similar technology, including baking, roasting and different wet processing systems.

4.2 Application of extrusion technology

Extrusion cooking technology is being increasingly applied worldwide to manufacture an ever expanding list of food products and feed materials, including snacks, breakfast cereals, pastas, texturized vegetable proteins (TVPs), pet foods, animal foods, instant beverage products and meat analogues and extenders (Table 4.1).

Moreover, an extruder can be operated with a low-moisture containing feed such that post-extrusion drying is optional or minimized. Furthermore, an extruder is a versatile machine; for example, an extruder can manufacture a breakfast cereal in the morning shift production, a meat analogue in the day shift, and expanded snacks in the night shift if appropriate hardware changes

Table 4.1 Brief list of extruded products

Product group	Product example	Reference
Grain products	Baby food, weaning food, cereal bar, bread crumb, instant porridge	Gopalakrishana and Jaluria (1992); Pinkaew, Wegmuller and Hurrell (2012)
Texturized plant protein	Meat substitute, soy granules	Moscicki (2011); Bhattacharya *et al.* (2009)
Modified starches	Speciality foods, raw material for convenience foods, enzyme-resistant starch foods	Smith (1992); Nehmer, Nobes and Yackel (2007)
Flavours	Encapsulated flavours	Camire (2000); Kollengode, Hanna and Cuppett (1996)
Dairy products	Sodium caseinate	Queguiner, Salou and Cheftel (1992); Manoi and Rizvi (2009)
Breakfast cereals	Corn/rice/wheat flakes, puffed products	Bouvier, Clextral and Firminy (2001)
Drinks	Beverage powder	Kazemzadeh (2001)
Muscle foods	Restructured fish/meat mince	Noguchi (1990); Choudhury and Gautam (2003)
Spice products and flavours	Sterilized/encapsulated spice powder	Schay (1975); Wojtowicz *et al.* (2010)
Confections	Chocolate, fruit gum liquorice	Muliji *et al.* (2003); Moscicki (2011)
Snacks	Ready-to-eat co-extruded and filled products, expanded flavoured snacks	Guy (2001); Altan, McCarthy and Maskan (2009); Meng *et al.* (2010); Seth and Rajamanickam (2012); Pilli *et al.* (2008)
Animal feed	Aquaculture feed, pet food, poultry feed	Williams (2000); Muthukumarapan (2011)
Baked goods	Flat bread, bread crumb	Moscicki (2011)
Pasta products	Oriental/quick cooking noodles, macaroni	Moscicki and Wojtowicz (2011); Zardetto and Rosa (2009)
Fats and oils	Stabilization of rice bran	Kim *et al.* (1987)

are made in addition to adjusting the extrusion operating conditions. It can also be operated with different types of raw materials, such as cereal/pulse flour, protein concentrates and oilseed cakes. It is thus expected that some more innovative extruded products having improved nutritional status and consumer acceptability will be available in the near future.

4.3 Description of an extruder

An extruder is a screw pump that contains a screw with flights and is encased in a barrel. The rotation of the screw inside the barrel makes the moist powdery

or granular material move forward and finally comes out of the die. The die can be considered as a restriction that is placed at the end of the extruder, having a diameter much small than that of the barrel. During the process of extrusion, the food material undergoes several unit operations such as mixing, conveying, shearing, heating and compaction. However, all these unit operations depend on several factors, such as the type and design of the extruder, the level of the operating variables (temperature at different regions of the barrel and at die, rotational speed of the screw, feed rate, etc.) and the characteristics of the feed (particle size, moisture content of the feed, protein/fat content, etc.). Nevertheless, the hardware of the extruder plays an important role in deciding the characteristics of the finished products. Details on different types of extruders and the components are available (Yacu, 2012; Harper, 1981a).

Extrusion of foods can be divided into two general categories, forming and cooking (Yacu, 2012). In the forming extrusion system, low-shear extruders are employed just to mix and shape to desired features. A marginal increase in temperature is expected while cooking is not the objective. The product application includes pasta, cold-formed snacks and other unexpanded or marginally expanded pellets. The forming extruder screw typically has a deep channel and is operated at a low speed. The forming extrusion is sometimes wrongly termed as 'cold extrusion'. Cold extrusion actually means a process when the whole operation is conducted at a low temperature (say 10 °C) while also maintaining a low die temperature for extrusion of muscle foods (Zuilichem, Janssen and Moscicki, 2011; Noguchi, 1990).

On the other hand, cooking extrusion applications normally utilize the medium and high shear single/twin-screw extruders. The source of energy is through viscous dissipation of mechanical energy, heat transfer through the barrel and by direct steam injection. The gap between the tip of the screw and barrel is kept at a minimum for improved shearing and heating. Both single- and twin-screw extruders have significant advantages and disadvantages; the specific advantages of a twin screw are better control of the extrusion process and product, and operation at lower pressures, while the disadvantages include the higher cost of the machine.

4.3.1 Type of extruder

Many kinds of extruders are used in the food industries and several designs are possible for a food extruder. The commonly used extruder consists of the flighted screw(s) or worm(s) rotating within a sleeve or barrel. Schematic drawings of the screw of a single- and a twin-screw extruder are shown in Figures 4.1 and 4.2, respectively. On the basis of the number of screws, extruders can be classified as single-screw or twin-screw extruders.

Single-screw extruder A single-screw extruder usually consists of three sections: feed, transition and metering zones (Figure 4.1). The extruder

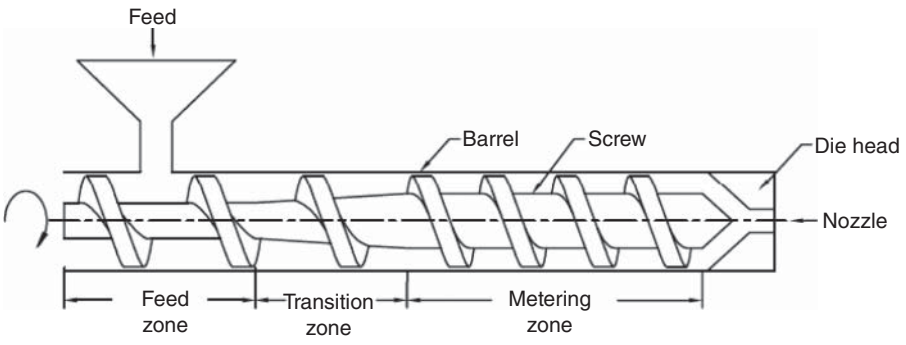

Figure 4.1 Single-screw extruder showing the screw profile

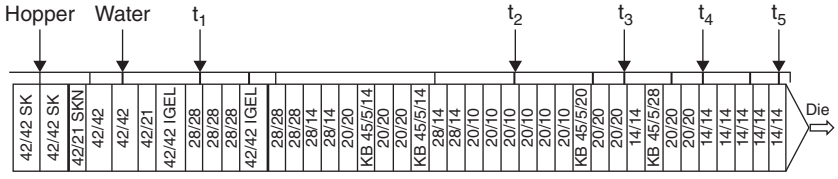

Figure 4.2 The screw profile of a twin-screw extruder (SK, SKN, conveying elements; IGEL, KB, mixing elements; L, left-handed elements; 42/42 = pitch/screw length; 45/5/14 = staggering angle of discs/number of discs/element length) (source: Moraru *et al.*, 2002. Reproduced with the permission of John Wiley and Sons, Ltd.)

screw sequentially conveys the foods forward (in the feed section) and heats the food ingredients to work them into a continuous plasticized mass (in the transition and metering sections) while rotating inside the barrel. The screw can be designed either as a single piece or as several small segments. The single-screw extruders are usually of five types, such as collet extruders (which operate with high shearing forces to induce rapid viscous dissipation of mechanical energy input for the production of crisp, expanded curl or collets), pasta extruders (having a deep flighted screw and operate at a low screw speed to produce pasta and macaroni), high pressure forming extruders (having a grooved barrel to prevent slip at the wall for the production of ready-to-eat snacks), high-shear cooking extruders (possessing a significant range of operating ability and being able to accept a wide range of feed moisture and ingredients with an ability to control the desired processing conditions for the production of pet foods, expanded cereals and snacks) and low-shear cooking extruders (operating with a moderate shear, high compression ratio and grooved barrel to enhance mixing for handling material with relatively low viscosity).

Twin-screw extrudes Twin-screw extruders are generally categorized according to the direction of screw rotation and the extent to which the screws intermesh. On the basis of rotation, these are known as counter-rotating and co-rotating twin-screw extruders. Twin-screw systems are considerably more complex in design than that of the single-screw extruder (Martelli, 1983). Twin-screw extruders are 1.5–3.0 times as expensive for a given throughput as compared to single-screw extruders. The food industries prefer co-rotating fully intermeshing twin-screw extruders because of several technological advantages, including ease of extruder operation and better control of the quality of the product.

4.3.2 Components of an extruder

Extruders are unique pieces of food processing equipment and a typical extruder consists of six major components. These are:

1. Feed assembly. This section invariably feeds to the extruder while optional functions are pre-conditioning, blending and moisturizing. The hopper bin is accompanied by a vibratory or screw feeder for continuous flow of the raw material to the extruder cooker, either by the principle of volumetric feeding or gravimetric feeding.
2. Extrusion barrel. The hollow cylindrical barrel is normally segmented for ease of assembly and disassembly. The interior surfaces of the barrel of a single screw contain small grooves and channels to increase friction. A removable sleeve, called the barrel liner, is often inserted within the barrels, which resist wear and can be replaced easily, if required. Heating elements are placed on the external barrel surface in addition to a jacket to allow the circulation of a medium for heating or cooling for temperature control. A venting facility for a twin-screw extruder may be used along with temperature probes at selected locations; inlet for water or steam is also provided in the barrel.
3. Extruder screw. The screw of a single-screw extruder can be divided into feed, compression and metering sections. The feed section has deeper flights and/or flights of a greater pitch. After moving away from the feed section, the material is compressed in the transition or the compression sections due to the presence of a tapered screw. The last zone (metering section) of the screw has shallow flights for increasing the shearing action leading to temperature increases.

   In the case of a twin-screw extruder (Figure 4.2), the root diameter of the screw(s) does not change in the barrel, unlike single-screw machines. The pitch of the screw here decreases towards the die and several elements including reverse pitch screw elements (RPSEs), mixing desks (MDs) or paddles, kneading block (KB) (a few mixing discs form a kneading block) and cut flights (or windows) are included in the screw profile primarily to

increase the residence time and/or shearing action. It is possible that the screws may be double- or even triple-start screws.

4. Extruder drive. An electrical motor, operating with a gear reduction facility, is employed to rotate the screw to provide the possibility of variation in the screw speed by using magnetic, electrical or mechanical controls. The ease of screw (or barrel) removal during jamming is also an essential feature.
5. Extrusion discharge. A number of shaped holes are provided in the die assembly or discharge section where the extrudates emerge from the extruder. It comprises a die or die insert, cutter, takeaway device and dryer/cooler. A variable-speed rotating knife is used to shape or cut the extruded product. The cut extrudates are taken away, usually by a belt or pneumatic conveyor for drying/cooling. A multipass belt-conveyer dryer or rotating drum dryer can cool or dry the sample to deliver a product to the post-extrusion system or packaging section.
6. Sensing, control and safety features. Sensors are provided to show the temperature at different barrel sections and die, pressure at die and torque during extrusion. Control of temperature is mandatory during extrusion. Safety cut-off levels are set on torque and pressure to avoid any potential damage to the extruder.

4.4 Selected extrusion technology

The technology of extrusion cooking has been applied for the production of many food products, of which pasta products, breakfast cereals, texturized vegetable protein (TVP), baby foods, confectionery products and expanded snacks are highly common. A generalized flowchart for the production of extruded products is shown in Figure 4.3 and some of the important extruded products (shown in Figures 4.4 and 4.5) are discussed.

4.4.1 Pasta products

Pasta products (e.g. dry macaroni, spaghetti, vermicelli and noodles) are now produced by employing high-capacity forming extruders. The target is to obtain a shaped product while a marginal temperature rise offers only a little cooking. Conventional pasta products need cooking for several minutes in boiling water while pre-cooked or instant pasta reduces the time of preparation at the consumer's end. The raw materials for pasta products are milled wheat (Kruger, Matsuo and Dick, 1996), water, egg (optional), salt and sometimes vegetable pieces/powder, protein rich sources, emulsifier (lecithin), glycerol monostearate, vitamins and minerals. The raw materials, after mixing and adjusting to a moisture content of about 30%, are shaped into various forms (thin round/rectangular cross-section, wheels, hollow tubes, curls, etc.) followed by drying in a controlled manner and packaged for market.

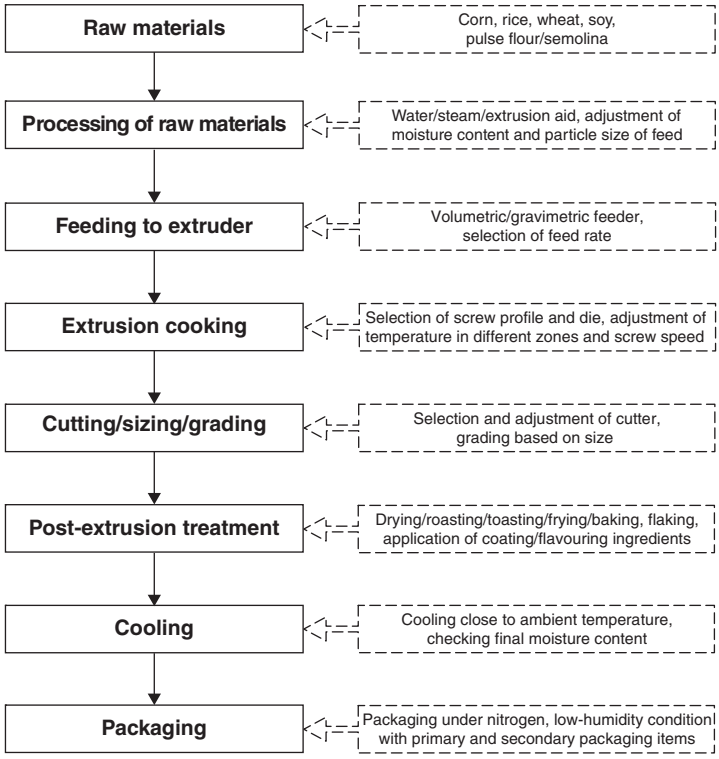

Figure 4.3 A generalized flowchart for extrusion cooking of foods

Figure 4.4 Pasta product in dried form (left) and after hydration (right) prepared from rice flour

Some improved pasta presses are equipped with a vacuum chamber to remove air bubbles from the pasta before extruding. If air is present in the mix prior to extrusion, small bubbles may be formed in the pasta, resulting in reduced mechanical strength of the finished product, increased oxidation of pigments and an undesirable white and chalky appearance. The traditionally employed bronze dies are now replaced with Teflon inserts, which improve

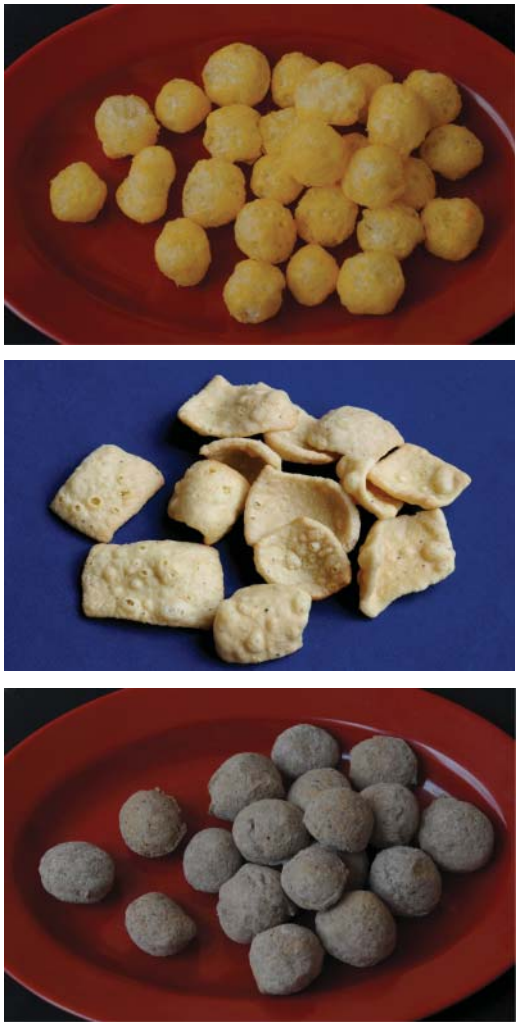

Figure 4.5 Various extruded products like expanded corn balls (top), extruded-toasted corn chips (middle) and texturized vegetable protein (TVP) (bottom). *See plate section for colour version*

the quality of the pasta and offer a smooth surface. Post-extrusion drying is a critical step affecting the quality of pasta; the reduction in moisture content to about 12% needs to be achieved with controlled conditions of drying temperature and relative humidity wherein too fast or too slow drying is to be avoided. The dried pasta product should be smooth without cracks and should have sufficient strength to retain integrity after cooking. The colour should be creamish or white, and not brownish. The solid loss during cooking should be minimum.

The price of pasta presses/extruders is much lower compared to extruder cookers of similar capacity. The design of such a pasta extruder, a control system and its maintenance are also simpler.

4.4.2 Breakfast cereals

Ready-to-eat (RTE) breakfast cereals are processed grain formulations suitable for human consumption without requiring further processing or cooking. Corn flakes are possibly the most common form of breakfast cereals (Fast, 1990). An increasingly wide range of breakfast cereals in attractive shapes are now available that are coated with various sweet formulations, enriched with additives such as filling (co-extruded pillows), food fibres like bran, micronutrients and other health benefiting ingredients.

The technology of extrusion cooking has been successfully applied since the 1970s to manufacture several breakfast cereals. The advantages of extrusion technology over the conventional method of corn flake production are the possibility of different shapes and uniformity of product size and shape.

Breakfast cereals can be categorized into traditional (hot) cereals that require further cooking or heating before consumption and ready-to-eat (cold) cereals that can be consumed directly or with the addition of milk. The steps in extruded flake production are pre-processing, mixing, extrusion cooking to obtain less expanded pellets, drying, cooling, tempering, flaking, toasting and packaging. For nonflaked breakfast cereals like balls/rings/curls, low-density expanded products are desirable instead of less expanded pellets. Variables that influence the extrusion process include screw speed, feed rate, barrel temperature, screw configuration, die geometry, moisture content of feed, die exit pressure and the residence time of feed inside the extruder. An appropriate modification of these process variables results in the required texture, density, size and shape of the product. Extruded breakfast cereals with multicereals, nuts and dry fruit pieces are gaining popularity. The products are usually fortified with minerals (iron, calcium, phosphorous, magnesium, zinc), fibres (bran, soluble dietary fibre), water-soluble and fat-soluble vitamins and sweet flavourings to improve the nutritional status. Actual processes in the production of breakfast cereals may vary to some extent depending on the type of cereal used. The cereals mostly used for the manufacture include corn, wheat, oat and rice. Different aspects of the manufacture of breakfast cereals employing conventional and extrusion cooking technology are available (Moscicki and Moster, 2011).

Extruded flakes differ from traditional flakes in that the extruded pellets are flaked in contrast to flaking of cooked grits. The extrudates leaving the extruder in a tube form are simultaneously filled with cream for developing cream-filled breakfast cereals. Later, they are flattened between two rollers followed by cutting into pieces in such a way as to seal both open ends. The post-extrusion treatments like toasting offer a crisp texture that stays longer

in milk while sweet coating using sugar, honey, chocolate and maltodextrin improves consumer acceptability. Corn flakes are toasted by placing them suspended in a hot air stream rather than using a baking surface. Carefully toasted breakfast cereals have the desired colour, moisture content and the attractive surface crispness due to the formation of a dry crust.

One or more of the cereal grains or milled fractions thereof are the major constituents of all breakfast cereals. The breakfast cereal ingredients may be classified (Caldwell and Fast, 1990) as (a) grains or grain products, (b) sweeteners, caloric or otherwise, (c) other flavouring or texturizing macroingredients, (d) microingredients for flavour and colour and (e) microingredients for nutritional fortification and extending shelf-life.

4.4.3 Texturized vegetable protein (TVP)

Several researches on the application of extrusion cooking and expander cooking technology of vegetable/oil seeds/legume protein-rich materials have made possible the manufacture of texturized vegetable protein (TVP), texturized plant protein (TPP) or meat analogue. Extrusion processing offers inactivation of the antinutritional factors contained in soybeans and legumes and improves product taste and denaturation of protein (Matyka *et al.*, 1996). Extrusion of texturized proteins is one of many successful applications of this unique cooking process. Extrusion-cooked texturized proteins include meat extenders in the form of chunks or small granular pieces. TVP serves as a protein-rich source for many people who wish to avoid meat and meat products due to religious and health reasons.

The textured vegetable proteins (TVPs) are produced through an extrusion system using slightly toasted defatted soy flour. This defatted soy flour usually meets the required characteristics of 45% protein content (minimum), 3.5% fibre (maximum), 1.5% fat (maximum) and protein dispersibility index (PDI) between 60 and 70. Soy flour allows controllable production of textured proteins in chunks. Other vegetable protein sources can also be used as raw materials for texturizing, and these include glandless cottonseed flour, rape seed or canola concentrates, defatted peanut flour, defatted sesame flour and soybean grits/flakes/meal/concentrates/isolates. The use and development of twin-screw extrusion cookers in the field of texturized proteins has increased the raw material specification range to include raw materials that include lower PDI ranges, higher fat and fibre levels and larger particle sizes (Kearns, 1988). The particle size requirements of defatted soy flour are of intermediate size. Very fine flour (below 40 μm) offers lumping during wetting while a very coarse sample (over 180 μm) requires the additional step of pre-moistening or pre-conditioning, and there is a chance that whole granules are visible in the finished product.

Calcium chloride ($CaCl_2$) between 0.5 and 2.0% is an effective additive in improving the textural integrity and surface smoothening of a TVP.

The addition of sodium alginate has also been reported to increase chewiness, water-holding capacity and density of a TVP. The other useful additives are soy lecithin (up to 0.4%), cysteine (between the 0.5 to 1% level) and bleaching agents such as hydrogen peroxide (between 0.25 and 0.5%) and titanium dioxide (between 0.5 and 0.75%).

Pre-conditioning before extrusion cooking considerably helps deactivation of growth inhibitors and significantly increases the efficiency of the process. Frequently, water or steam is directly injected into the barrel during the production of TVPs.

Extruded meat analogues in the form of soy steaks, cutlets and other meat analogues have proved successful in the market owing to their fibrous structure and nutritional value, similar to that of meat products. Textured proteins of vegetable or animal origin can be processed into meat-like extruded compounds by two methods: dry and wet texturization (Moscicki, 2011). A spongy texture is the characteristic of the dry texturization process. The dried product is rehydrated prior to final use. Wet extruded products do not need any rehydration as they are processed near the final moisture content of the product.

The processing of texturized protein can be done on both single- and twin-screw extruders. The mechanism of texturization includes cross-linking and alignment of proteins at die (Harper, 1981b); it is a function of the time, temperature, and shearing and moisture history (Harper *et al.,* 1978). The meat extenders, when ground to a particle size simulating hamburger, are used at 25–30% levels in dishes such as pizza, hamburgers, meat loaf and meat sauces. Chunk-style products are effectively used in soups, stews, meat pies, dry soup mixes and oriental dishes.

4.4.4 Snack foods

Extruded snacks have the greatest potential for growth among the snack food category as the extrusion technology can provide the opportunity to process a variety of products by changing a minor ingredient and processing condition on the same machine; different shapes, sizes, structural, textural and sensory attributes and colour of snack foods are possible to manufacture by using an extruder. The consumer demand for 'good for health' snack foods leads to the elusive search for something unique that attracts the consumer, and extrusion technology can meet these requirements to produce healthy snacks from health benefiting ingredients.

The corn curls/balls are expanded using a collet extruder, followed by drying/baking/frying and flavouring. The filled snacks require special attachments like the co-extrusion facility wherein twin-screw extruders are better suited.

There are many ways to classify the snacks. Snack manufacturers use three main terms to identify the snacks (Seker, 2011). The first-generation snacks are all the natural/conventional products; examples include ready-to-eat nuts, potato chips and popped popcorn. The second-generation snack is based on

the single ingredient snacks and simple shaped products like corn tortilla chips and other directly expanded snacks. The third-generation snacks (also called half-product or pellets) comprise multi-ingredient formed snacks and pellets, and include cheese balls, corn curls, corn sticks, etc.

The extrusion cooked pellets are the semi-finished products that are formulated from a variety of mixtures of starchy materials like wheat, rice, oat and corn flour, pulses, oilseed meals and processed potato. It is desirable that the formulation recipe contains a minimum of 60% starch in order to obtain snacks with a crispy texture (Keller, 1989). The various type of additives that can be included in the formulation are thermally/chemically modified starch and fragmented protein products of animal origin (shrimp, fish, poultry, beef, cheese and powdered milk) and vegetable origin (seeds of leguminous plants, meals and concentrates), basically used to increase the protein content and raise the nutritional value of the products.

A two-stage extrusion process is sometimes used to manufacture the half-products. The first extruder (called as a gelatinizer or G extruder) performs the gelatinization or melting of the crystalline structures in starch granules along with an adequate quantity of moisture and raised temperature. The hot fluid expands at the die, flashes off some water and is rapidly partially cooled. It then enters the feed port of the forming extruder (called as the F extruder). After extrusion, the half-products lose some water vapour and may have moisture contents of 20–25%. They are dried further to around 10% moisture content. They can be expanded into low-density foam structures at a later time. However, this process can be substituted by employing a pre-conditioner for the initial cooking (gelatinization) followed by a forming operation by a twin-screw extruder. The example of such products includes flaked flavoured snacks.

4.5 Post-extrusion treatment

The purpose of post-extrusion treatment is primarily to bring the product to a safe moisture level, thus improving the sensory acceptance with particular reference to texture, flavour and taste. The other objectives are the avoidance of an undesirable flavour and a change in the size and shape so that the finished product is improved from the technological point of view and consumer liking. On some occasions, the extrudates are converted to a ready-to-eat form by employing the post-extrusion treatment from the ready-to-cook form. The post-extrusion treatment may include roasting/toasting, drying, frying, baking, flavouring and coating, or a combination of these unit operations. For example, the texturized vegetable protein, made from extrusion cooking of defatted soy flour, is dried to a safe moisture content of about 6% by employing a rotary inclined drum dryer or a multipass continuous belt dryer. The purpose of baking, frying or roasting/toasting is to improve

the product in terms of texture, flavour and taste while reducing the moisture content of the finished product; a good amount of cooking is also performed in these processing steps. The processing involving flaking, flavouring and coating improves the acceptance of the product. Batch and continuous flavour applicators, sprinklers and coating devices can be employed. This is the crucial step for the acceptance of expanded ready-to-eat snacks. The process of toasting/roasting is especially important for breakfast cereals wherein the crisp surface texture, improved integrity in milk and appealing flavour are developed. Several ingredients are used during post-extrusion processing (Bhattacharya, 2011; Seighman, 2001). These are salt, sugar, spice, colour, oleoresin, oil, protein isolates, honey, maltodextrin, malt, chocolate, edible gum, corn syrup, liquid glucose, organic acid (e.g. citric acid), antioxidant, wax, fruit and vegetable powders, minerals and vitamins, and different flavours (e.g. cheese, fruit, fish, spice, vanilla, rock salt and monosodium glutamate).

The extruded pellets are passed through a small distance between a pair of counter-rotating rolls at a temperature of approximately 45 °C to produce thin extruded flakes. The slight roughness of the roll surface and the hard or dry surface of the pellet provide friction and flow between the rolls without fracture of the pellet (Maskan and Altan, 2011). Gelatinization, moisture content and strength of the pellet also provide flow between the rolls and integrity of the pellets during passage through the rolls. High moisture causes sticky flakes that adhere to the rolls. After the pellets pass through the rolls for flaking, the resulting flakes may curl up (Fast, 1990).

Crispness and blister of flakes are provided by drying and toasting of flakes; this is particularly important for flaked breakfast cereals. Flakes are held at a temperature of about 150 °C to reduce the moisture content further and then held at a temperature of 200–300 °C, depending on the types of cereal toasting for a short duration (Miller, 1994). Flakes are toasted in a rotary toasting oven or in a continuous hot-air oven followed by cooling of the products.

Products can be coated with syrup and flavour additives. Hot and concentrated sugar solution is pumped to be sprayed through a nozzle on to extrudates. Replacing some part of sucrose with invert sugars reduces the crystallization of sugars. The coated extrudates are re-dried after coating. Extruded products are frequently coated or flavoured with various liquid or powdery ingredients to improve their appeal to consumers. The use of encapsulated flavour seems to be an alternative (Yuliani *et al.*, 2004). Dry additives like nuts and dry fruits can be added and then the final products are packaged to be transported to stores.

Starch with a high amylose content causes harder and denser extrudates that absorb milk slowly, but starch with a high amylopectin content provides expanded extrudates. On the other hand, the modification of starch with cross-linking reduces with water absorption and a solubility index of starch extrudates (Seker and Hanna, 2006, 2005).

4.6 Quality characteristics of product

The quality of extruded products is difficult to define precisely, but refers to the degree of excellence of a food and includes all the characteristics that have a significance in determining the degree of acceptability. Different methods and measurements are used for quality control of foods, which includes shape and size, sensory characteristics, expansion indices, colour, texture, density, physicochemical properties and structural characteristics. These quality factors depend on the composition of the product, process variables, expected deteriorative reactions, packaging used and shelf-life of the product. The quality characteristics of extruded products can be broadly grouped into the following categories:

(a) During extrusion: torque, specific mechanical energy (SME) input and rheological status (Hsieh. Peng and Huff, 1990).
(b) Physical, physicochemical and sensory characteristics of the product: expansion ratio, water absorption and water-holding capacities, rheological status of the product in water, bulk density, textural attributes (Ravi, Roopa and Bhattacharya, 2007), porosity, moisture content, extent of gelatinization and retrogradation, mechanical acoustic attributes, sensory evaluation, flavour and appearance/colour.
(c) Nutritional characteristics: nitrogen solubility index (NSI), protein and fat contents, anti-nutritional factors, enzyme activity, protein digestibility index, amino acid balance, microbial status and starch digestibility (Harper, 1981b).
(d) Structural/microstructural features: type/number/size and shape of air cells, thickness of the cell wall and their alignment (Agbisit *et al.*, 2007), fibre formation and alignment in meat analogues (Ranasinghesagara *et al.*, 2009). The microstructure of an expanded corn ball is different from corn-soy extrudates in terms of cell size and wall thickness.

The intensity and extent of changes after extrusion treatment may be defined by various quality determinants. The selection of appropriate quality attributes depends on the final product. For example, when evaluating the pasta, a number of parameters are taken into account including the minimum preparation time (cooking time), water absorption index, water solubility index, degree of starch gelatinization, cooked weight, the amount of organic ingredients passing into the water during cooking (cooking loss), and texture and sensory characteristics (Li and Vasanthan, 2003; Smewing, 1997). On the other hand, physical properties (like density and expansion) and textural, sensory and microstructural features are more important for the ready-to-eat extruded snacks. In general, for a starch-rich product, the extent of gelatinization determines the intensity of heat treatment in a given material and degree of cooking achieved. The extruded food products may not display 100%

gelatinized starch because a small part reacts with other food ingredients and creates insoluble and unavailable complexes (Pagani, 1986).

The nutritional aspects are of general interest in all extruded products. The effects of extrusion cooking on nutritional quality are ambiguous. Beneficial effects include inactivation of antinutritional factors, undesirable enzymes and microorganisms, gelatinization of starch and increased soluble dietary fibre contents. However, Maillard reactions between protein and sugars reduce the nutritional value of the protein, decrease in heat-sensitive vitamins and changes in proteins and amino acid profile also occur due to extrusion cooking (Singh, Gamlath and Wakeling, 2007).

4.7 Equations related to food extrusion

The unit operation of extrusion cooking has been modelled using several equations; a significant portion of them is based on an empirical approach.

Several extrusion processing systems are judged by torque during extrusion. Further, the operation of extruders is often limited by a cut-off torque value; this acts as a safety parameter. The torque ($T\%$) during extrusion is usually obtained from the digital display of the extruder. The total specific mechanical energy (SME) input during extrusion can be estimated using the following equation (Hsieh, Peng and Huff, 1990):

$$\text{SME} = \frac{\text{Rpm of screw (run)}}{\text{Rpm of screw (rated)}} \times \frac{\%\ \text{Torque (run)}}{100} \times \frac{\text{Motor power (rated)}}{\text{Production capacity}} \tag{4.1}$$

The rheology of dough is an important index that affects the process of extrusion. The apparent viscosity of dough (η) during extrusion is shown by the following equation (Harper, Rhodes and Wanniger, 1971).

$$\eta = C_1\, \dot{\gamma}^{C_2}\, e^{C_3/T}\, e^{C_4 M} \tag{4.2}$$

Here, T is the temperature of the sample (K), $\dot{\gamma}$ is the shear rate (s^{-1}), M is the moisture content and C_1, C_2, C_3 and C_4 are empirical constants.

The net volumetric flow rate (Q) from an extruder can be expressed as follows (Das, 2005):

$$Q = Q_D - Q_P - Q_L \tag{4.3}$$

where Q_D is the contribution from drag flow, Q_P is the pressure flow and Q_L is the leakage flow; the last two parameters oppose the drag flow and hence possess negative signs. The drag flow is caused by the rotation of the extruder screw, which carries the material towards the die end. The pressure and leakage flows should be as minimum as possible. The pressure flow is caused by the development of high pressure at the die end and the leakage flow is the flow that occurs between the tip of the screw and the barrel.

The flow rate from the extruder is also equal to the flow rate from the die. Assuming that the flow through the die is laminar, the value of Q can be obtained from the Hagen-Poiseulli equation as follows (Das, 2005):

$$Q = \frac{\pi \ d^4 \ P}{128 \ l \ \mu_d} \tag{4.4}$$

Here, d and l are the diameter and length of the die, respectively, P is the gauge pressure at the die and μ_d is the apparent viscosity at the die.

As an extrusion cooking system is frequently approached in an empirical manner (black box modelling), a second-order polynomial is used to relate extrusion variables (moisture content of feed, screw speed, feed rate, temperature, etc.) with product characteristics or response functions (such as density, expansion ratio, etc.). Usually, an experimental design in the coded (x) levels of variables is employed to reduce the number of experiments. The response functions (Y_{ijk}) are shown as

$$y_{ijk} = b_0 + \sum_{i=1}^{n} b_i x_i + \sum_{\substack{i=1 \\ i \le j}}^{n} \sum_{j=1}^{n} b_{ij} x_i x_j + \varepsilon_{ijk} \tag{4.5}$$

In Equation (4.5), the number of variables is denoted by n while i, j and k are integers. The coefficients of the polynomials are represented by b_0, b_i and b_{ij}, and ε_{ijk} is the random error. When $i < j$, b_{ij} represents the interaction effects of the variables x_i and x_j (Myers, 1971).

In addition, modelling studies are available that relate to the wear of an extruder, flow in different screw profiles, residence time distribution, scale-up and changes in physical/chemical/thermal/nutritional properties due to extrusion cooking.

The extruded product quality (target product parameters) is dependent not only on the raw material used for extrusion but also on the design of the machine and on the operating conditions of the extruder. Thus, extrusion processing of foods is a complex system, mostly due to the presence of a large number of process, system and design variables; the use of an artificial neural network (ANN) appears to be useful.

4.8 Present status

Extrusion cooking of food is now more than 40 years old and continues to grow with new areas of application, use of nonconventional raw materials, changes in the design of machines and the development of new products. It is also expected that extrusion technology can thus offer itself as an alternative processing method of some of the traditional products (appearing to be difficult to mechanize at present) in a better hygienic way.

The other expected specific technological advancement includes dual extrusion and co-extrusion technology, where the flow rate of the two extruders has to be matched. Conventional extruder cookers are used in food industries for producing extruded snacks, breakfast cereals, texturized proteins and confectionery products. However, lower equipment costs, lower maintenance costs with lower wear costs, higher throughput, better control of the process, easier dismantling and cleaning of the extruder and the extrusion of feed with higher levels of fat and sugar are the targets of producers and equipment producers. A turbo-extruder as a special pump working on the basis of friction has been developed and patented to achieve some of the targets (Heinz, 1996). The turbo-extrusion process operates with an extremely low residence time of 8 s compared to a convectional twin-screw extruder, which operates at a residence time of 20–60 s. It has been claimed that the turbo-extrusion process can provide better product stability and texture and can handle feed material with higher fat and sugar contents (Seker, 2011).

Extrusion with supercritical fluids is another promising area for future application. The injection of a supercritical fluid like carbon dioxide into an extruder has been attempted by Ferdinand, Clark and Smith (1992). The production of expanded extrudates is possible by eliminating the effect of steam as carbon dioxide can act as a blowing agent instead of using a process to flush off moisture (Seker, 2011). Hence, it is possible to achieve expansion of extrudates at lower temperatures. Smaller cells with thick cell walls are possible for supercritical fluid-assisted extruded products.

Existing areas with added research can also provide the industrial production of (a) high moisture extrusion of muscle protein, (b) development of biodegradable products, (c) use of an extruder cooker as a continuous reactor, (d) replacing commonly used processing systems (such as baking and frying technology) by extrusion technology, (e) developing traditional foods through extrusion, and (f) creating new foods with unique sensory features. Mathematical modelling of the changes during extrusion cooking and automation in extrusion processing are also the areas of interest to researchers and machinery manufacturers. It is expected that this proven technology will bring some more new food products having innovative features and nutritional superiority.

References

Agbisit, R., Alavi, S., Cheng, E., Herald, T. and Trater, A. (2007) Relationships between microstructure and mechanical properties of cellular cornstarch extrudates. *Journal of Texture Studies*, **38**, 199–219.

Altan, A., McCarthy, K.L. and Maskan, M. (2009) Effect of extrusion cooking on functional properties and *in vitro* starch digestibility of barley-based extrudates from fruit and vegetable by-products. *Journal of Food Science*, **74**, E77–E86.

Barron, C., Valle, D.G., Colonna, P. and Vergnes, B. (2002) Energy balance of low hydrated starches transition under shear. *Journal of Food Science*, **67**, 1426–1437.

Benbow, J. and Bridgwater, J. (1993) *Paste Flow and Extrusion*. Clarendon Press, University of Michigan, Michigan, USA.

Bhattacharya, S. (2011) Raw materials. In: *Advances in Extrusion Technology* (Maskan, M. and Altan, A., eds). CRC Press, Taylor and Francis Group, Boca Raton, Florida, USA, pp. 69–85.

Bhattacharya, S., Ramesh, T., Baby Latha, R., Appu Rao, A.G. and Prakash, V. (2009) Method for preparing multipurpose ready-to-use high-protein soy granules. US Patent 7,541,057 B2.

Bouvier, J.M., Clextral, S.A. and Firminy, L. (2001) Breakfast cereals. In: *Extrusion Cooking Technologies and Applications* (Guy, R., ed). CRC Press (Taylor and Francis), Boca Raton, Florida, USA, pp. 133–160.

Caldwell, E.F. and Fast, R.B. (eds) (1990) *Breakfast Cereals and How They Are Made*. American Association of Cereal Chemists, Inc., St Paul, Minnesota, USA, pp. 15–42.

Camire, M.E. (2000) Chemical and nutritional changes in food during extrusion. In: *Extruders in Food Applications* (Riaz, M.N., ed). CRC Press (Taylor and Francis), Boca Raton, Florida, USA, pp. 127–142.

Chang, Y.K. and Wang S.S. (1998) *Advances in Extrusion Technology*. Technomic Publishing Co., Cornell University, Cornell, New York, USA.

Choudhury, G.S. and Gautam, A. (2003) Hydrolyzed fish muscle as a modifier of rice flour extrudate characteristics. *Journal of Food Science*, **68**, 1713–1721.

Das, H. (2005) *Food Processing Operations Analysis*. Asian Book Private Limited, New Delhi, India.

Ferdinand, J.M., Clark, S.A. and Smith, A.C. (1992) Structure formation in extrusion-cooked starch-sucrose mixtures by carbon dioxide injection. *Journal of Food Engineering*, **16**, 283–291.

Fast, R.B. (1990) Manufacturing technology of ready-to-eat cereals. In: *Breakfast Cereals and How They Are Made* (Fast, R.B. and Caldwell, E.F., eds). American Association of Cereal Chemists, Inc., St Paul, Minnesota, USA, pp. 15–42.

Frame, N.D. (ed.) (1993) *Technology of Extrusion Cooking*. Chapman and Hall, New York, USA.

Gopalakrishana, S. and Jaluria, Y. (1992) Modelling of starch gelatinisation in a single-screw extruder. In: *Food Extrusion Science and Technology* (Kokini, J.L., Ho, C.T. and Karwe, M.V., eds). Marcel Dekker, New York, USA, pp. 3–19.

Guy, R. (ed.) (2001) *Extrusion Cooking Technologies and Applications*. Woodhead Publishing Limited, Abington, Cambridge, England.

Harper, J.M. (1981a) *Extrusion of Foods*, vol. **1**. CRC Press, Boca Raton, Florida, USA.

Harper, J.M. (1981b) *Extrusion of Foods*, vol. **2**. CRC Press, Boca Raton, Florida, USA.

Harper, J.M., Rhodes, T.P. and Wanniger, L.A. Jr, (1971) Viscosity model for cooked cereal dough. *Chemical Engineering Programs*, **67**, 40–43.

Harper, J.M., Cummings, D.J., Kellerby, J.D., Tribelhorn, R.E., Jansen, G.R. and Maga, J.A. (1978) Evaluation of low-cost extrusion cookers for use in LDCs (Annual Report), Colorado State University, USA.

Heinz, S. (1996) Cooker-extruder apparatus and process for cooking-extrusion of biopolymers. US Patent 5, 567,463.

Hsieh, F., Peng, I.C. and Huff, H.E. (1990) Effects of salt, sugar and screw speed on processing and product variables of corn meal extruded with a twin-screw extruder. *Journal of Food Science*, **55**, 224–227.

Janssen, L.P.B.M. (1978) *Twin Screw Extrusion*. Elsevier Scientific Publication Co., New York, USA.

Janssen, L.P.B.M. (2004) *Reactive Extrusion Systems*. Taylor and Francis. The University of Groningen, The Netherlands.

Jorge, C., Alvarez, M. and Rao, M. (2011) Industrial application of extrusion for development of snack products including co-injection and pellet technologies. In: *Advances in Extrusion Technology* (Maskan, M. and Altan, A., eds). CRC Press (Taylor and Francis), Boca Raton, Florida, USA.

Jowitt, R. (ed.) (1984) *Extrusion Cooking Technology*. Elsevier Science Publishing Co., Inc., New York, USA.

Kazemzadeh, M. (2001) Baby foods. In: *Extrusion Cooking: Technologies and Application* (Guy, R., ed.). Woodhead Publishing Ltd, Cambridge, Emgland, pp. 182–199.

Kearns, J.P. (1988) *Extrusion of Texturized Proteins*. American Soybean Association, Wenger International, Inc., Kansas City, Missouri, USA.

Keller, L.C. (1989) Method for producing expanded, farinaceous food product. US Patent 4, 869,911.

Kim, C. J., Byun, S. M., Cheigh, H. S. and Kwon, T. W. (1987) Optimization of extrusion rice bran stabilization process. *Journal of Food Science*, **52**, 1355–1357.

Kokini, J.L., Ho, C.T. and Karwe, M.V. (eds) (1992) *Food Extrusion Science and Technology*. Marcel Dekker, New York, USA.

Kollengode, A.N.R., Hanna, M.A. and Cuppett, S. (1996) Volatiles retention as influenced by method of addition during extrusion cooking. *Journal of Food Science*, **61**, 985–990.

Kruger, J.E., Matsuo, R. and Dick, J. (1996) *Pasta and Noodle Technology*. American Association of Cereal Chemists, Inc., St Paul, Minnesota, USA.

Li, J. and Vasanthan, T. (2003) Hypochlorite oxidation of field pea starch and its suitability for noodle making using an extrusion cooker. *Food Research International*, **36**, 381–386.

Manoi, K. and Rizvi, S.S.H. (2009) Physicochemical changes in whey protein concentrate texturized by reactive supercritical fluid extrusion. *Journal of Food Engineering*, **95**, 627–635.

Martelli, F.G. (1983) *Twin-Screw Extruders: A Basic Understanding*. Van Nostrand Reinhold Co, New York, USA.

Maskan, M. and Altan, A. (eds) (2011) *Advances in Food Extrusion Technology*. CRC Press (Taylor and Francis), Boca Raton, Florida, USA.

Matyka, S., Mościcki, L., Jaśkiewicz, T. and Pokorn, J. (1996) Extrusion-cooked faba bean-rapeseed concentrate in broiler chicks diets. *Scientia Agriculturae Bohemica*, **27**(3), 211–220.

Meng, X., Threinen, D., Hansen, M. and Driedger, D. (2010) Effect of extrusion conditions on system parameters and physical properties of a chick pea based snack products. *Food Research International*, **43**, 650–658.

Mercier, C., Linko, P. and Harper, J.M. (eds) (1989) *Extrusion Cooking*. American Association of Cereal Chemists, St Paul, Minnesota, USA.

Miller, R.C. (1994) Die and cutter design in food extrusion. American Association of Cereal Chemists Short Course, 17–19 June, Leuven, Belgium.

Moraru, C. I., Lee, T.C., Karwe, M.V. and Kokini, J.L. (2002) Phase behaviour of a meat-starch extrudates illustrated in a state diagram. *Journal of Food Science*, **67**, 3026–3032.

Moscicki, L. (2011) Processing of full fat soybeans and textured vegetable proteins. In: *Extrusion-Cooking Techniques Applications, Theory and Sustainability* (Moscicki, L., ed.). Wiley-VCH Verlag GmbH, Weinheim, Germany, pp. 119–126.

Moscicki, L. and Moster, A. (2011) Production of breakfast cereals. In: *Extrusion-Cooking Techniques and Applications: Theory and Sustainability* (Moscicki, L., ed.), Wiley-VCH Verlag GmbH, Weinheim, Germany, pp. 65–80.

Moscicki, L. and Wojtowicz, A. (2011) Raw materials in the production of extrudates. In: *Extrusion-Cooking Techniques and Applications: Theory and Sustainability* (Moscicki, L., ed.). Wiley-VCH Verlag GmbH, Weinheim, Germany, pp. 45–60.

Muliji, N.C., Miquel, M.E., Hall, L.D. and Mackley, M.R. (2003) Microstructure and mechanical property changes in cold extruded chocolate. *Food and Bioproducts Processing*, **81**, 97–105.

Muthukumarapan, K. (2011) Extrusion of pet foods and aquatic feeds. In: *Advances in Extrusion Technology* (Maskan, M. and Altan, A., eds). CRC Press (Taylor and Francis), Boca Raton, Florida, USA, pp. 237–255.

Myers, R.H. (1971) *Response Surface Methodology*. Allyn and Bacon, Boston, Massachusetts, USA, pp. 61–106.

Nehmer, W.L, Nobes, G.A. and Yackel, W.C. (2007) Production of enzyme-resistant starch by extrusion. US Patent US2007/0275155A1.

Noguchi, A. (1990) Recent research and industrial achievements in extrusion cooking in Japan. In: *Processing and Quality of Foods*, vol, 1. *High Temperature/Short Time (HTST) Processing Guarantee for High Quality Food with Long Shelflife* (Zeuthen, P., Cheftel, J.C., Eriksson, C., Gormley, T.R., Linko, P. and Paulus, K., eds). Elsevier Applied Science, London, England, pp. 1.203–1.213.

Pagani, M.A. (1986) *Pasta and Extrusion Cooked Foods*. Elsevier Applied Science Publishers, London, England.

Pilli, T.D., Carbone, B.F., Derossi, A., Fiore, A.G. and Severini, C. (2008) Effects of operating conditions on oil loss and structure of almond snacks. *International Journal of Food Science and Technology*, **43**, 430–439.

Pinkaew, S., Wegmuller, R. and Hurrell, R. (2012) Vitamin A stability in triple fortified extruded, artificial rice grains containing iron, zinc and vitamin A. *International Journal of Food Science and Technology*, **47**, 2212–2220.

Queguiner, C., Salou, E.D.C.C. and Cheftel, J.C. (1992) Application of extrusion cooking to dairy products: preparation of fat analogue by microcoagulation of whey proteins. In: *Food Extrusion Science and Technology* (Kokini, J.L., Ho, C.T. and Karwe, M.V., eds). Marcel Dekker, New York, USA, pp. 373–386.

Ranasinghesagara, J., Hsieh, F.-H., Huff, H. and Yao, G. (2009) Laser scanning system for real-time mapping of fiber formations in meat analogues. *Journal of Food Science*, **74**, E39–E45.

Ravi, R., Roopa, B.S. and Bhattacharya, S. (2007) Texture evaluation by uniaxial compression of some snack foods. *Journal of Texture Studies*, **38**, 135–152.

Riaz, M.N. (2000) Practical consideration in extrusion cooking. In: *Extruders in Food Applications* (Riaz, M.N., ed.). CRC Press (Taylor and Francis), Boca Raton, Florida, USA, pp. 149–165.

Schay, R. (1975) Natural flavours. In: *Fenaroli's Handbook of Flavour Ingredients*, vol. 1 (Furia, T.E. and Bellanca, N., eds). CRC Press, California, USA, pp. 271–495.

Seighman, B. (2001) Snack food seasonings. In: *Snack Foods Processing* (Lusas, E.W. and Rooney, L.W., eds). Technomic Publications, Lancaster, Pennsylvania, USA, pp. 495–516.

Seker, M. (2011) Extrusion of snacks, breakfast cereals and confectioneries. In: *Advances in Extrusion Technology* (Maskan, M. and Altan, A., eds). CRC Press (Taylor and Francis), Boca Raton, Florida, USA, pp. 168–208.

Seker, M. and Hanna, M.A. (2005) Cross-linking starch at various moisture content by phosphate substitution in an extruder. *Carbohydrate Polymers*, **59**, 541–544.

Seker, M. and Hanna, M.A. (2006) Sodium hydroxide and trimetaphosphate levels affect properties of starch extrudates. *Industrial Crops Products*, **23**, 249–255.

Seth, D. and Rajamanickam, G. (2012) Development of extruded snacks using soy, sorghum, millet and rice blend – a response surface methodology approach. *International Journal of Food Science and Technology*, **47**, 1526–1531.

Singh, S., Gamlath, S. and Wakeling, L. (2007) Nutritional aspects of food extrusion: a review. *International Journal of Food Science and Technology*, **42**, 916–929.

Smewing, J. (1997) Analyzing the texture of pasta for quality control. *Cereal Foods World*, **42**, 8–12.

Smith, A.C. (1992) Studies on physical structure of starch based materials in extrusion cooking process. In: *Food Extrusion Science and Technology* (Kokini, J.L., Ho, C.T. and Karwe, M.V., eds). Marcel Dekker, New York, USA, pp. 573–618.

Steven, M.J. and Covas, J.A. (1995) *Extruder Principles and Operation*. Chapman and Hall, London, England.

Williams, M.A. (2000) Interrupted-flight expanders-extruders. In: *Extruders in Food Applications* (Riaz, M.N., ed.). CRC Press (Taylor and Francis), Boca Raton, Florida, USA, pp. 63–78.

Wojtowicz, E., Wojtasiak, R.Z., Adamiec, J., Wasowicz, E. and Przygonski, K. (2010) Odor active compounds content in spices and their microencapsulated powders measured by SPME. *Journal of Food Science*, **75**, S441–S445.

Yacu, W.A. (2012) Extruder selection, design, and operation for different food applications. In: *Advances in Extrusion Technology* (Maskan, M. and Altan, A., eds). CRC Press (Taylor and Francis), Boca Raton, Florida, USA, pp. 23–67.

Yuliani, S., Bhandari, B., Rutgers, R. and D'Arcy, B. (2004) Application of microencapsulated flavour to extrusion product. *Food Reviews International*, **20**, 163–185.

Zardetto, S. and Rosa, M.D. (2009) Effect of extrusion process on properties of cooked, fresh egg pasta. *Journal of Food Engineering*, **92**, 70–77.

Zuilichem, D.J.V., Janssen, L.P.B.M. and Moscicki, L. (2011) Engineering aspects of extrusion. In: *Extrusion-Cooking Techniques: Applications, Theory and Sustainability* (Moscicki, L., ed.). Wiley-VCH Verlag GmbH, Weinheim, Germany, pp. 25–43.

5
The Process of Gelling

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5.1 Introduction

A liquid material called 'sol' becomes a solid material called 'gel' by the process known as 'gelling'. The diversified applications of gels for convenience are available in several fields, including various products belonging to the food and pharmaceutical industries. However, common examples of natural gels start from the cytoplasm of cells to the soft pulpy part of a tender coconut, while man-made gels include jam, jelly and a number of gelled food products. A typical structured or textured product like a gel is a preferred form of use compared to the liquid-like 'sol' behaviour. Indeed, the process of gelling is unique and needs a proper understanding to obtain a gel of the required attributes.

The commercial importance of gelled products is undeniable in a world where convenience of use is a critical factor. The other factor is the creation of a specialized texture in the so-called gelled products; its importance lies in developing different analogues like fruit/meat/crab/shrimp imitation products, some of which are already popular while others are expected to be a commercial success in the near future.

Gels are created out of 'sol'; the latter contains selected ingredients, which on further processing (called the gelling process) forms the solid-like 'gel', which is a typical semi-solid or viscoelastic substance exhibiting both solid and liquid characteristics. The ingredients for developing food gels are usually from proteins or hydrocolloids, which possess the gel-forming ability under appropriate conditions. The important factors for gelling are concentration of the gelling agent, pH, temperature and time of thermal processing, the presence of cations and co-solutes, etc.

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The general raw materials that possess the gelling ability are proteins (gelatin, albumin, soy and whey protein isolates/concentrates, etc.) and hydrocolloids (pectin, gellan, agar, xanthan, carrageenan, etc.). Each of these materials requires specific conditions to form a gel; the formed gels from different raw materials have typical characteristics, such as texture, syneresis, appearance, chewing ability, colour, opacity/transparency and taste. Thus, the success of a good gelled product depends on careful selection of the gelling agent and subsequent processing conditions.

A number of commercial gel products are available off the shelf. The examples are various traditional products like jam/jelly/marmalade/jujube, confectionaries, different imitation products like mock meat, etc. A few book chapters (Foegeding, 2005; Walstra, 2003), several reviews on food gels and the gelling process (Banerjee and Bhattacharya, 2012; Oakenful, 1987) and many research articles and patents are available. The structural and functional characteristics, and applications of hydrocolloids have also been discussed (Laaman, 2011; Phillips and Williams, 2009; Hoefler, 2004). The present chapter thus focuses on the diversified process of gelling along with examples, processing conditions, characterization and the possibility of new type of gels.

5.2 Classification of gels

Gels may be classified in several ways, of which classification based on thermoreversibility (Figure 5.1a) and on the source of gelling agent (Figure 5.1b) is convenient for understanding. Many formed gels such as gelatin, agar, xanthan and gellan show thermoreversibility, meaning that these gels can be converted to the 'sol' state by increasing its temperature, and vice versa. On the other hand, only a few gels, like alginate and high methoxy pectin gels, are thermoirreversible in nature, indicating that once they are formed, they cannot be brought back to the liquid state by heating (Williams and Phillips, 2000). However, both thermoreversible and thermoirreversible gels have certain advantages/drawbacks and hence their applications are different.

Gels may be categorized based on the source of the gelling agents. The major sources are plant, animal, algal and microbial sources of which pectin, gelatin and starch are the oldest gelling agents that are still in use for developing different traditional products and commercially important items in several countries (Table 5.1). The high-protein-containing gelling agents are derived from soy bean, gelatin, alginate, caseinate and whey protein in the form of powder, concentrate and isolate. Several applications of these gels are possible, starting from the conventional gels like jam, jelly, coated products to unique items like a dehydrated film.

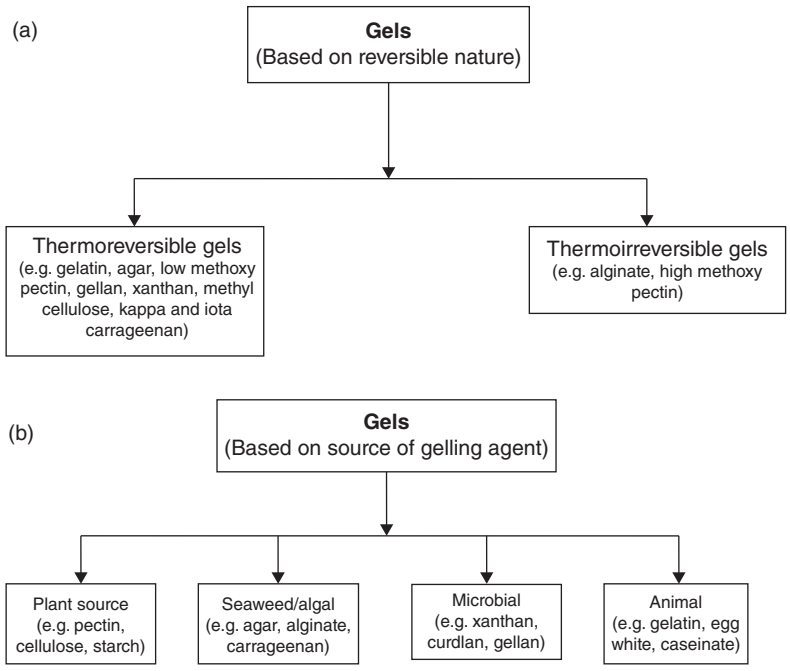

Figure 5.1 Classification of gels based on (a) thermoreversibility and (b) source of the gelling agent

Based on temperature dependence of the elastic modulus, polysaccharide gels may be classified into four categories (Nishinari and Zhang, 2004):

1. Cold set gels like agarose, carrageenan and gellan, which form a gel on cooling the dispersion.
2. Heat set gels like some cellulose derivatives [methyl cellulose (MC) and hydroxy propyl methyl cellulose (HPMC)], curdlan and konjac glucomannan, which form a gel on heating the dispersion.
3. Re-entrant gels like xyloglucan, which form gels at intermediate specific temperature ranges and remain in the sol state at higher and lower temperatures outside this temperature range.
4. Inverse re-entrant gels like a mixed dispersion of methyl cellulose and gelatin, which form a gel at higher and lower temperatures and stay in the sol state in the intermediate temperature range.

Gels belonging to the first class are mostly thermoreversible and have been studied extensively, although the gelation mechanism has not completely been clarified at the molecular level. The other three classes have not been elucidated so well. Some polysaccharides like MC, HPMC or curdlan form

Table 5.1 Categories of food gels made from different sources

Gelling agent	Application	Other ingredients	Reference
Plant sources			
Pectin	Jam, jelly, pudding, yoghurt, marmalade, jujubes	Sugar, fruit pulp	Williams and Phillips (2000); Oakenfull (1987)
Starch, starch derivates and starch-rich sources	Pudding, rice gel	Sugar, egg, pulse flour, milk, konjac glucomannan	Alka and Bhattacharya (2008); Charoenrein *et al.* (2011)
Soy protein	Convenience food gel	Soy protein isolate (SPI)	Bhattacharya and Jena (2007)
Cellulose and cellulose derivatives	Salad dressings and deserts	Oil, sugar	Agoub and Morris (2008); Williams and Phillips (2000)
Algal sources			
Agar	Used as laxative, vegetarian gelatin substitute, jelly, Japanese desserts such as *anmitsu*	Sugar, acid	Armisen and Galatas (2000); Stanley (2006)
Carrageenan	Gel with immobilized cells/enzymes	Other hydrocolloids	Aguilera and Stanley (1999); Williams and Phillips (2000)
Alginate	Jelly, gelation with divalent cations, cell immobilization and encapsulation, appetite suppressant, coating of fruit/fruit pieces, edible films and food coatings	Enzymes, microbes	Roopa and Bhattacharya (2008); Grant *et al.* (2005); Rhim (2004)
Microbial source			
Xanthan	Salad dressings and sauces, helps to stabilize the colloidal oil and solid components against creaming by acting as an emulsifier and texture modifier	Guar gum, alginate, sugar, fruit pulp	Peressini, Pin and Sensidoni (2011); Kayacier and Dogan (2006); Sutherland (2007); Doublier and Cuvelier (2006)
Gellan	Coating on fruit pieces, texturized fruit	Alginate, fruit pulp, gellan	Tapia *et al.* (2007); Papageorgiou and Kasapis (1995)
Curdlan	Low-calorie and emulsion type food	Fat is replaced by curdlan	Nakao, Okur and Tawada (1994)
Animal sources			
Gelatin/collagen	Gelling, water binding, film-forming, reduces surface tension, microencapsulation	Other hydrocolloids	Gómez-Guillén *et al.* (2011)
Egg protein	Gelling, thickening, emulsifying, foaming, colouring	Other hydrocolloids	Williams and Phillips (2000)

a gel on heating; however, konjac glucomannan (KGM) forms a gel on heating only in the presence of alkali, which removes acetyl groups. Some of these heat-set gels are thermoreversible (MC or HPMC gel and low-set gel of curdlan) while others (KGM gel and high-set gel of curdlan) are thermoirreversible (Nishinari and Zhang, 2004). In addition, based on their composition, gels are also categorized as polysaccharide gels (such as those formed by alginate, carrageenan, pectin, agar, gellan, starch, etc), protein gels (like those formed by ovalbumin, casein, soy protein and gelatin) and mixed gels (protein-polysaccharide). Mixed biopolymer gels, especially protein-polysaccharide combinations, provide a wide range of textural properties and hence are extensively used in food formulations. The best known example is *k*-carrageenan-milk protein gel, which is used in the preparation of various dairy desserts (Puvanenthiran *et al*., 2003). Depending on the number of gelling agents used, gels can be single-component or multicomponent gels. Multicomponent gels often offer the advantage of synergistic interactions thereby improving the gel properties. Various multicomponent gels are in use in foods like carrageenan-starch-milk proteins in dairy desserts (Verbeken *et al*., 2006, 2004), gellan-gelatin in dessert jellies (Sworn, 2000) and gellan-modified starch in glazed bakery products (Sworn, 2000).

Another type of gel is the sheared gel/fluid gel. Many polysaccharides like agar and *k*-carrageenan form a microgel sample if a suitable shear field is applied to the sample during gelation. Such microgels are called sheared gels or fluid gels (Norton, Frith and Ablett, 2006). Several traditional products are available where the usage of hydrocolloids has been known for several decades.

5.3 Gelling process

The process of gelling usually has four steps, such as (a) hydration of the gelling agent, (b) mixing with other ingredients, (c) heating of mixed 'sol' and (d) cooling of the 'sol' in moulds to allow the gel to set (Figure 5.2). However, the process varies depending on the gelling agent and the ingredients to be added. Temperature plays an important role which needs to be carefully monitored and controlled to get gels of the desired size, shape, texture and sensory attributes. It is obvious that these gels contain high levels of moisture so their shelf-life is short. Refrigeration/freezing of the formed gels can, however, increase their shelf-life to some extent. On the other hand, if the formulations are appropriately selected and/or final gels are dried to a safe value of water activity, the products can be converted into a ready-to-eat form having a moderate shelf-life of 3 to 6 months without requiring low-temperature storage.

To make the gels on a commercial scale, the required machineries are mixing devices and heating equipments to obtain 'sol' and moulding. Several unique shapes such as a candle, stick, animals, fruits, etc., are possible depending on the mould employed.

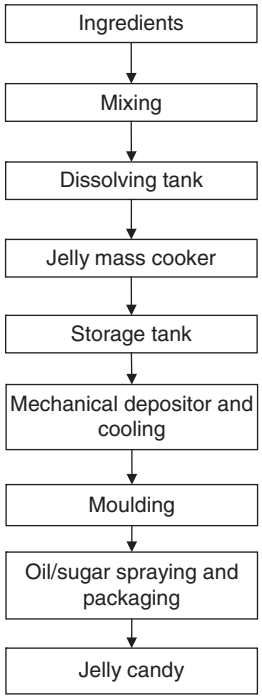

Figure 5.2 Flowchart for the production of jelly candy

5.4 Mechanism of gel formation

The stability of the gel is mainly influenced by the intermolecular forces between the gelling agents in the presence of a dispersion medium. The intermolecular forces may be weak in nature, such as hydrogen bonds, electrostatic forces, Van der Waals forces, hydrophobic interactions, etc. On the other hand, a strong force like the disulfide bond (covalent bond) is responsible for the formation of protein gels (Oakenfull, 1987). However, a combination of weak intermolecular forces gives a stable gel by cross-linking the molecules together. Two or more polymeric molecules are cross-linked to form a 'junction zone' and many such zones give rise to a gel. Thus, the gelation process is controlled by the formation of junction zones and the intermolecular forces involved in it. It is also worth noting that these cross-linkages are permanent until a stress is applied externally (Oakenfull, 1987).

The structural and conformational changes occurring during gelling are complex in nature and are affected by various factors like temperature, the presence of ions, pH, the inherent structure of gelling agents, the presence of other additives and co-solutes, etc. The dispersion medium (mainly water) is held in the three-dimensional polymeric network of the gelling agent(s).

The basis for the three-dimensional network of the gel is the formation of 'junction zones' by aggregation of primary interchain linkages. The three important proposed mechanisms for explaining the phenomenon of gelation of hydrocolloids are ionotropic gelation, cold-set gelation and heat-set gelation (Oakenfull, 1987).

In ionotropic gelation, the cross-linking of polymeric chains with ions (mostly cations) leads to gel formation or gelation. This can be carried out by two main techniques such as (a) diffusion setting, in which the diffusion of ions into the hydrocolloid dispersed medium leads to gelation, and (b) internal gelation, in which the inactivated ions are dispersed initially in the hydrocolloid dispersion followed by its activation by changing the pH, leading to gel formation. The latter technique is advantageous as in the diffusion setting the gels formed are inhomogeneous in nature with a soft core and firm surface (Oakenfull, 1987).

Cold-set gelation is characterized by the formation of polymeric networks, initially by heating the hydrocolloid dispersed sample and later by cooling it. Interchain helices are formed from the segments of individual chains on cooling the dispersion, leading to the formation of the three-dimensional network in the gel. Examples include agarose, gellan, etc. (Banerjee and Bhattacharya, 2012).

Heat-set gelation occurs by unfolding or expansion of the native structures of the hydrocolloid (like starch) on heating. The native structures undergo subsequent rearrangement to form a stable network in the presence of heat (Burey *et al.*, 2008).

An agar gel and a gellan gel are shown in Figure 5.3. The significant difference in the physical parameter like transparency/opacity is worth mentioning.

Gels can also be prepared by aeration, where the physical properties and the material characteristics vary significantly after aeration. Figure 5.4 shows the conventional and aerated gellan gels; the transparent nature of the gellan gel is considerably lost on aeration.

5.5 Methods for characterization of gels

The process of transformation of sol to the state of gel can be monitored by various methods, such as heat flow data, structural features including microscopic observations, physical characteristics like opacity/transparency, sensory attributes and mechanical and rheological properties (Banerjee and Bhattacharya, 2012). Among these methods, the rheological properties of sol as well as gel are of prime importance. Table 5.2 shows the rheological measurements while Table 5.3 indicates the other type of tests; both of these tests need to be linked with a sensory assessment of the samples.

The sensory scores of trained panelists are equally important because it is the consumer or the end user who ultimately decides the use and/or role of

Figure 5.3 Agar (top) and gellan (bottom) gels

Figure 5.4 Conventional (left) and air-incorporated (right) gellan gels. *See plate section for colour version*

Table 5.2 Methods for the rheological measurement of gel characteristics (source: Banerjee and Bhattacharya 2012. Reproduced with permission of Taylor & Francis Ltd.)

Nature of test	Type of measurement	Instrument used	Measurement parameters	Applications	References
Fundamental tests	Compression	Texture measuring system	Modulus of elasticity, Poisson's ratio	Surimi gel	Kim *et al.* (2006); Hamann and MacDonald (1992); Walstra (2003)
	Stress relaxation	Texture measuring system	Residual stress, relaxation time	Gellan gels	Morris (1986)
	Creep	Controlled stress rheometer	Shear modulus, creep compliance	Soy and gelatin gels	Kamata and Kinsella (1989); Chronakis *et al.* (1995)
	Oscillation	Controlled stress rheometer	Storage modulus (G'), loss modulus (G''), phase angle (δ), complex modulus and viscosity	Viscoelastic characterization of rice, soy gels, mixed gels	Jena and Bhattacharya (2003); Kim *et al.* (2006); Bhattacharya and Jena (2007); Keogh *et al.* (1995)
Empirical	Puncture force	Texture measuring system	Puncture characteristics	Characterization of rice gel	Jena and Bhattacharya (2003); Kim *et al.* (2006)
	Compression	Texture measuring system	Peak force, firmness, compression energy	Measurement of gel quality and gel strength	Kim *et al.* (2006); Smewing (1999)
Imitative	Texture profile analysis (TPA)	Texture measuring system	Hardness, fracturability, adhesiveness, springiness, cohesiveness, chewiness	Food gels	Pons and Fiszman (1996)

these types of products in everyday life. Sensory parameters such as hardness, cohesiveness, springiness, stickiness and overall acceptability can be assessed and used for evaluating the commercial feasibility of the product.

Common instruments, capable of measuring fundamental and empirical rheological properties of fluid and semi-solid foods, are commercially

Table 5.3 Other methods for the measurement of gel characteristics (source: Banerjee and Bhattacharya 2012. Reproduced with permission of Taylor and Francis Ltd.)

Type of measurement	Instrument used	Measurement parameters	Applications	Reference
Structural characterization	Differential scanning calorimeter (DSC)	Heat flow	Gellan and polyvinyl alcohol blend film	Sudhamani *et al.* (2003)
	X-ray diffraction	Particle size analysis	Nano delivery system in food	Luykx *et al.* (2008)
	Colorimeter	Colour measurement	Gellan edible films	Leon *et al.* (2008)
Microscopic characterization	Light microscope (LM)	Area of the granules	Tapioca starch gel	Vittadini *et al.* (2006)
	Scanning electron microscope (SEM)	Structural arrangement of components	The number, area and location of particles in gel	Moritaka *et al.* (2003)
	Transmission electron microscope (TEM)	Structural distribution of constituents	Characteristic studies of mixed gel	Aguilera and Stanley (1999)
	Atomic force microscope (AFM)	Structure of the molecules	Structural characteristics of nanoparticles	Luykx *et al.* (2008)
Molecular characterization	Nuclear magnetic resonance (NMR)	Conformational changes on gelation	Structural features of the constituents	Saito *et al.* (1995)
	Fourier transform and infrared(FTIR)	Molecular structure	Infrared spectra of the components	Sudhamani *et al.* (2003)
	FT-Raman; near-infrared resonance	Molecular characterization	Functional characteristics of pectins	Wilats *et al.* (2006)
Proximate analysis	Vacuum oven	Moisture	Moisture estimation	Leon *et al.* (2008)
	Atomic absorption spectroscopy	Mineral content	Chitosan and whey protein isolate based model system	Laplante, Turgeon and Paquin (2005)
	Kjeldhal apparatus	Protein	Whey protein – cassava starch gel	Aguilera and Stanley (1999)
	DNS method	Carbohydrate	Sugar content of the gel	Aguilera and Baffico (1997)

available. Costs vary tremendously from the inexpensive glass capillary viscometer to a very expensive rheometer capable of measuring dynamic properties like oscillation, creep recovery and normal stress characteristics. The formed gels are often tested in compression (between parallel plates), shearing or torsion. The common routine empirical/imitative tests include puncture or penetration and texture profile analysis.

Some liquid foods at a high concentration of solids and food doughs show both viscous and elastic properties. The measurement of viscoelasticity is usually complex in nature and can be divided into two broad classes. The first, generally termed 'fundamental tests', measures properties that are inherent to the material and do not depend on the geometry of the test sample, the conditions of loading or on the apparatus. Examples of these properties are modulus of elasticity, Poisson's ratio, relaxation time and shear modulus. The other class, the empirical or imitative tests, is used to determine properties such as puncture force, stress relaxation and texture profile analysis (TPA).

The elastic modulus is the ratio of stress to strain on a material, wherein stress is equal to the force per unit area and strain is the observed deformation divided by the original length of the material. The fundamental tests measure well-defined rheological or specific viscoelastic properties. Fundamental tests are usually time consuming and need more sophisticated and costly instruments. However, they offer universally acceptable values and conventionally assume (a) a small applied strain, (b) the material is continuous, isotropic (exhibiting the same physical properties in every direction) and homogeneous and (c) the test sample is uniform and of regular shape. The parameters determined from fundamental tests are Young's modulus of elasticity, shear modulus, bulk modulus, Poisson's ratio, creep compliance, storage modulus, loss modulus, phase angle, etc. Though complicated, the fundamental tests are considered as one of the best techniques for quality control and product development purposes, particularly for viscoelastic or semi-solid materials like sol and gel.

5.6 Mathematical models

The rheology of gels can be characterized by different methods. Among them, the axial compression of a cylindrical-shaped sample between parallel plates is very common. During compression, the height of the sample decreases and its diameter increases. Thus, the commonly used engineering or apparent compressive strain ($\epsilon_{\mathbf{E}}$) can be calculated by

$$\epsilon_{\mathbf{E}} = \frac{\Delta h}{h_0} \tag{5.1}$$

where h_0 is the height of the sample at time $t = 0$ and Δh represents the change in height due to compression. If the cylindrical shape of the sample is retained

during compression, the true, natural or Hencky's compressive strain ($\epsilon_{\mathbf{T}}$) is

$$\epsilon_{\mathrm{T}} = \ln\left(\frac{h_0}{h_0 - \Delta h\,(t)}\right) \tag{5.2}$$

The engineering or apparent compressive stress (σ_{E}) can be obtained from the force ($F(t)$) and the initial cross-sectional area (A_0) of the sample:

$$\sigma_{\mathrm{E}} = \frac{F(t)}{A_0} \tag{5.3}$$

The true compressive stress (σ_{T}) is calculated from the following equation, where r_0 is the initial radius of the sample:

$$\sigma_{\mathrm{T}} = \frac{F(t)(h_0\text{-}\Delta h(t))}{\pi r_0^2 h_0} \tag{5.4}$$

Young's modulus is equal to the ratio of the true stress and true strain (up to the elastic limit) and is an important index to judge the elastic property of the gel samples. The relation between true strain and true stress can be given by a power law model:

$$\sigma_{\mathrm{T}} \;= k\,(\epsilon_{\mathrm{T}})^n \tag{5.5}$$

The empirical constant k represents the stiffness and the power index n is called the degree of concavity; it is the deviation from linearity as indicated by Hooke's law. If the degree of concavity is > 1, this indicates strain hardening while strain softening/weakening is expressed for values < 1.

In addition, when dealing with structured materials as gels and colloidal systems, the useful theory is probably the 'gel theory' or Winter theory (Gabriele, de Cindio and D'Antona, 2001), dealing with the evaluation of the sol–gel transition point. It is worth mentioning here that the critical concentration of the gel forming ingredient as well as the gelling temperature and time are important factors that dictate the gelling process. This theory characterises the gels, and gives some useful tools to evaluate with good approximation the 'gel point', where the liquid–solid phase transition occurs, by using simple rheological tests. The relaxation modulus follows a power law type equation:

$$G(t) = At^{-m} \qquad \text{for } \lambda_0 < t < \infty \tag{5.6}$$

where m is the relaxation exponent, A represents the gel strength and λ_0 is a characteristic shortest time for the crossover to small-scale dynamics.

Gels, being a typical semi-solid material, also follow the general principles of viscoelastic materials. Thus, the spring-dashpot models have been used to characterize the solid (represented by a spring) and liquid (represented by a dashpot) properties of gel samples, or several combinations of spring and dashpot, connected in series or parallel.

5.7 Conclusions

Gelling is a unique process where complex phenomena like interactions within a hydrocolloid and water molecule, simultaneous changes in molecular structure and the effects of pH and temperature play the vital roles. The process of gel formation and the various hydrocolloids used have a wide scope of study considering the development of newer products with unique sensory and functional characteristics. The multicomponent gel systems, where more than one hydrocolloid are used to form a gel, is gaining more interest in recent times. In addition, nutrients carrying gels and air-incorporated gels possess a good future. The available mathematical models prove to be useful in predicting the process parameters and product characteristics. The use of gel-forming ingredients like proteins and hydrocolloids and their combinations in food systems is of significant importance to obtain a product that satisfies the end users. At the international level, the regulations in the use of gelling agents in food are governed by the Codex Alimentarius Commission (CAC) and Joint Expert Committee on Food Additives (JECFA). The conclusions that can be derived from these legislations are that these agents are safe for use in food products under permitted limits while it is also mandatory to follow the laws of the country where such foods are expected to be consumed.

References

Agoub, A. A. and Morris, E. R. (2008) Particulate rheology and acid-induced gelation of oxidised cellulose. *Carbohydrate Polymers*, **71**, 416–427.

Aguilera, J. M. and Baffico, P. (1997) Structure–mechanical properties of heat induced whey protein/cassava starch gels. *Journal of Food Science*, **62**, 1048–1066.

Aguilera, J. M. and Stanley, D. W. (1999) Food structuring. In: *Microstructural Principles of Food Processing and Engineering* (Aguilera, J. M. and Stanley, D. W., eds). Chapman and Hall, New York, USA, pp. 185–249.

Alka, K. and Bhattacharya, S. (2008) Gelling behavior of rice flour dispersions at different concentrations of solids and time of heating. *Journal of Texture Studies*, **39**, 231–251.

Armisen, R. and Galatas, F. (2000) Agar. In: *Handbook of Hydrocolloids* (Phillips, G. O. and Williams, P. A., eds). Woodhead Publishing Limited and CRC Press LLC, Cambridge, England/Boca Raton, Florida, USA, pp. 20–39.

Banerjee, S. and Bhattacharya, S. (2012) Food gels: gelling process and new applications, *Critical Reviews in Food Science and Nutrition*, **52**, 334–346.

Bhattacharya, S. and Jena, R. (2007) Gelling behaviour of defatted soybean flour dispersions due to microwave treatment: textural, oscillatory, microstructural and sensory properties. *Journal of Food Engineering*, **78**, 1305–1314.

Burey, P., Bhandari, B.R., Howes, T. and Gidley, M.J. (2008) Hydrocolloid gel particles: formation, characterization and application. *Critical Reviews in Food Science and Nutrition*, **48**, 361–377.

Charoenrein, S., Tatirat, O., Rengsutthi, K. and Thongngam, M. (2011) Effect of konjac glucomannan on syneresis, textural properties and the microstructure of frozen rice starch gels. *Carbohydrate Polymers*, **83**, 291–296.

Chronakis, I.S., Kasapis, S., Richardson, R.K. and Doxastakis, G. (1995) Characterisation of a commercial soy isolate by physical techniques. *Journal of Texture Studies*, **26**, 371–389.

Doublier, J. L. and Cuvelier, G. (2006) Gums and hydrocolloids: functional aspects. In: *Carbohydrates in Food* (Eliasson, A.-C., ed.). Taylor & Francis Group, LLC, New York, USA, pp. 232–272.

Foegeding, A.E. (2005) Rheology, structure and texture perception in food protein gels. In: Food Colloids: *Interactions, Microstructure and Processing*. The Royal Society of Chemistry, Cambridge, England, pp. 3–15.

Gabriele, D., de Cindio, B. and D'Antona, P. (2001) A weak gel model for foods. *Rheologica Acta.* **40**, 120–127.

Gómez-Guillén, M.C., Giménez, B., López-Caballero, M.E. and Montero, M.P. (2011) Functional and bioactive properties of collagen and gelatin from alternative sources: a review. *Food Hydrocolloids*, **25**, 1813–1827.

Grant, S.C., Celper, S. Gauffin-Holmberg, I., Simpson, N.E., Blackband, S.J. and Constantinidis, I. (2005) Alginate assessment by NMR microscopy. *Journal of Material Science: Materials in Medicine*, **16**, 511–514.

Hamann, D.D. and MacDonald, G.A. (1992) Rheology and texture properties of surmi based food. In: *Surmi Technology* (Lanier, T.C. and Lee, C.M., eds). Marcel Dekker, New York, USA, pp. 429–500.

Hoefler, A. (2004) *Hydrocolloids – Practical Guides for the Food Industry*. Eagan Press Handbook Series, Eagan Press, St Paul, Minnesota, USA.

Jena, R. and Bhattacharya, S. (2003) Viscoelastic characterization of rice gel. *Journal of Texture Studies*, **34**, 349–360.

Kamata, Y. and Kinsella, J.E. (1989) A comparison of creep phenomena in food protein gels. *Journal of Food Science*, **54**, 170–172.

Kayacier, A. and Dogan, M. (2006) Rheological properties of some gums–salep mixed solutions. *Journal of Food Engineering*, **72**, 261–265.

Keogh, M.K., Laine, K.I. and O'Connor, J.F. (1995) Rheology of sodium caseinate–carrageenan mixtures. *Journal of Texture Studies*, **26**, 635–652.

Kim, H.-J., Decker, E.A. and McClements, D.J. (2006) Preparation of multiple emulsions based on thermodynamic incompatibility of heat-denatured whey protein and pectin solutions. *Food Hydrocolloids*, **20**, 586–595.

Laaman, R.T. (2011) *Hydrocolloids in Food Processing*. Blackwell Publishing Ltd and Institute of Food Technologists, Iowa, USA.

Laplante, S., Turgeon, S.L. and Paquin, P. (2005) Effect of pH, ionic strength, and composition on emulsion stabilizing properties of chitosan in a model system containing whey protein isolate. *Food Hydrocolloids*, **19**, 721–729.

Leon, P.G., Lamanna, M.E., Gerschenson L.N. and Rojas, A.M. (2008) Influence of composition of edible films based on gellan polymers on l-(+)-ascorbic acid stability. *Food Research International*, **41**, 667–675.

Luykx, D.M.A.M., Peters R.J.B., van Ruth, S.M. and Bouwmeester, H. (2008) A review of analytical methods for the identification and characterization of nano delivery systems in food. *Journal of Agricultural and Food Chemistry*, **56**, 8231–8247.

Moritaka, H., Kimura, S. and Fukuba, H. (2003) Rheological properties of matrix–particle gellan gum gel: effects of calcium chloride on the matrix. *Food Hydrocolloids*, **17**, 653–660.

Morris, E.R. (1986) Rheology of hydrocolloids. In: *Gums and Stabilizers for the Food Industry* (Philips, G.O., Williams, P.A. and Wedlock, D.J., eds)., IRL Press, Oxford, England, pp. 57–77.

Nakao, Y., Okur, Y. and Tawada, T. (1994) Emulsion-type food. US Patent 5,360,624A.

Nishinari, K. and Zhang, H. (2004) Recent advances in the understanding of heat set gelling polysaccharides. *Trends in Food Science and Technology*, **15**, 305–312.

Norton, I.T., Frith, W.J. and Ablett, S. (2006) Fluid gels, mixed fluid gels and satiety. *Food Hydrocolloids*, **20**, 229–239.

Oakenfull, D. (1987) Gelling agents. *Critical Reviews in Food Science and Nutrition*, **26**, 1–25.

Papageorgiou, M. and Kasapis. S. (1995) The effect of added sucrose and corn syrup on the physical properties of gellan–gelatin mixed gels. *Food Hydrocolloids*, **9**, 211–220.

Peressini, D., Pin, M. and Sensidoni, A. (2011) Rheology and breadmaking performance of rice – buckwheat batters supplemented with hydrocolloids. *Food Hydrocolloids*, **25**, 340–349.

Phillips, G.O. and Williams, P.A. (2009) *Handbook of Hydrocolloids*, 2nd edn. Woodhead Publishing Limited, Cambridge, England.

Pons, M. and Fiszman, S.M. (1996) Instrumental texture profile analysis with particular reference to gelled systems. *Journal of Texture Studies*, **27**, 597–624.

Puvanenthiran, A., Goddard, S.J., Mekinnon, I.R. and Augustin, M.A. (2003) Milk-based gels made with κ-carrageenan. *Journal of Food Science*, **68**, 137–141.

Rhim, J.-W. (2004) Physical and mechanical properties of water resistant sodium alginate films. *LWT-Food Science and Technology*, **37**, 323–330.

Roopa, B.S. and Bhattacharya, S. (2008) Alginate gels: I. Characterization of textural attributes. *Journal of Food Engineering*, **85**, 123–131.

Saito, H., Shimizu, H., Sakagami, T., Tuzi, S. and Naito, A. (1995) Conformation and dynamics of polysaccharide gels as studied by high resolution solid state NMR. In: *Magnetic Resonance in Food Science* (Belton, P.S., Delgadillo, I., Gil, A.M. and Webb, G.A., eds). Royal Society of Chemistry, London, England, pp. 227–271.

Smewing, J. (1999) Hydrocolloids. In: *Food Texture: Measurement and Perception* (Rosenthal, A. J., ed.). Aspen Publication, Gaithersburg, Maryland, USA, pp. 282–296.

Stanley, N.F. (2006) Agars. In: *Food Polysaccharides and Their Application*, 2nd edn. (Stephen, A.M., Phillips, G.O. and Williams, P.A., eds). CRC Press, Boca Raton, Florida, USA, pp. 217–238.

Sudhamani, S.R., Prasad, M.S. and Udaya Sankar, K. (2003) DSC and FTIR studies on gellan and polyvinyl alcohol (PVA) blend films. *Food Hydrocolloids*, **17**, 245–250.

Sutherland, I.W. (2007) Biotechnology of microbial polysaccharides in food. In: *Food Biotechnology*, 2nd edn (Shetty, K., Paliyath, G., Pometto, A. and Levin, E., eds). CRC Press, Taylor & Francis Group, Philadelphia, Pennsylvania, USA, pp. 193–220.

Sworn, G. (2000) Xanthan gum. In: *Handbook of Hydrocolloids* (Phillips, G.O. and Williams, P.A., eds). Woodhead Publishing Limited, New York, USA, pp. 103–115.

Tapia, M.S., Rojas-Graü, M.A., Rodríguez, F.J., Ramírez, J., Carmona, A. and Martin-Belloso, O. (2007) Alginate and gellan-based edible films for probiotic coatings on fresh-cut fruits. *Journal of Food Science,* **72**, 190–196.

Verbeken, D., Thas, O. and Dewettinck, K. (2004) Textural properties of gelled dairy desserts containing κ-carrageenan and starch. *Food Hydrocolloids*, **18**, 817–833.

Verbeken, D., Bael, K., Thas, O. and Dewettinck, K. (2006) Interactions between κ-carrageenan, milk proteins and modified starch in sterilized dairy desserts. *International Dairy Journal*, **16**, 482–488.

Vittadini, E., Carini, E. and Barbanti, D. (2006) The effect of high pressure and temperature on the macroscopic, microscopic, structural and molecular properties of tapioca starch gels. In: *Water Properties of Food, Pharamaceutical and Biological Materials* (del Pilar, B.M., Welti-Chanes, J., Lillford, P.J. and Corti, H.R., eds). Taylor & Francis Group, LLC, Philadelphia, Pennsylvania, USA, pp. 471–483.

Walstra, P. (2003) Soft solids. In: *Physical Chemistry of Foods*. Marcel Decker, Inc., New York, USA, pp. 683–771.

Wilats, W.G.T., Knox, J.P. and Mikkelsen, J.D. (2006) Pectin: new insights into an old polymer are starting to gel. *Trends in Food Science and Technology*, **17**, 97–104.

Williams, P.A. and Phillips, G.O. (2000) *Handbook of Hydrocolloids*. Woodhead Publishing Limited, Cambridge, England.

6

Thermal Food Preservation Techniques (Pasteurization, Sterilization, Canning and Blanching)

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6.1 Introduction

The use of heat for thermal processing to inactivate microorganisms has been one of the most widely used methods of food preservation for more than a century and has contributed significantly to the nutritional well-being of much of the world's population. The processing technologies most commonly used in the food industry consist of in-line pasteurization and sterilization of liquids through the use of heat exchangers and hold tubes prior to filling into containers, and in-container pasteurization or sterilization (canning) consisting of heating filled and sealed food containers in pressurized retorts (autoclaves or 'pressure cookers'). Blanching is a relatively mild form of heat treatment applied to fresh foods prior to preservation processes to release entrapped air and inactivate enzymes. This chapter is intended to describe the processing technologies used in the food industry to accomplish these different types of heat treatments.

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6.2 Pasteurization and sterilization

Thermal processing in the food industry covers the broad area of preservation technology in which heat treatments are used to inactivate microorganisms to accomplish either commercial sterilization or pasteurization. Sterilization of foods is normally accomplished by what is commonly called 'canning' to preserve the safety and wholesomeness of ready-to-eat foods over long terms of extended storage at normal room temperature. These are known as shelf-stable foods. Pasteurization is normally used to extend the limited shelf-life of refrigerated foods. Both processes make use of heat treatments to inactivate microorganisms. However, they differ widely with respect to the classification or type of microorganism targeted, the range of temperatures that must be used, and the type of equipment systems capable of achieving such temperatures.

6.2.1 Pasteurization

Purpose of pasteurization Thermal pasteurization is a relatively mild heat treatment given to foods with the purpose of destroying selected vegetative species of microorganisms, such as the many pathogens that cause food-borne illness, as well as to accomplish inactivation of enzymes. Pasteurization does not eliminate all vegetative microorganisms, nor does it eliminate more heat-resistant spore-forming bacteria. Therefore, pasteurized foods are not shelf-stable and must be stored under refrigeration and/or with modified-atmosphere packaging, which slow the growth of microorganisms that still remain viable in the product and will ultimately cause spoilage. Depending on the type of product, the shelf-life of pasteurized foods could range from several days (milk) to several weeks or more (fruit juices). Because only relatively mild heat treatment is involved, the sensory quality and nutritive value of the food are minimally affected. The severity of heat treatment used (time and temperature) and the length of shelf-life achieved depends on the nature of the product, pH, heat resistance of the target microorganisms, sensitivity of the product quality to heat damage, and the method of heating.

Pasteurization equipment systems Liquid products that are capable of being pumped through tubular pipe lines, like milk and fruit juices, are thermally pasteurized by heating/holding/cooling while the products are flowing through a system of heat exchangers and hold tubes. These systems deliver a high-temperature short-time (HTST) process sufficient to inactivate target microorganisms while causing minimal heat damage to product quality. Unpasteurized liquid products are held cold in 'raw product' refrigerated tanks until ready to be pasteurized. As raw product is pumped through the heat exchanger system, the product temperature is quickly raised to the

required pasteurization temperature as it flows through the 'heating' heat exchanger, sent through an insulated holding tube to receive the required length of time at the pasteurizing temperature, and quickly cooled and chilled through the 'cooling' heat exchanger to the refrigerated temperature for storage in a 'pasteurized product' refrigerated tank until ready for filling.

Pasteurizing temperatures fall in the range of 60–80 °C, well below the boiling point of water at atmospheric pressure. Since water is typically used as the heat exchange medium in these systems, there is no need to operate under pressure. Either plate heat exchangers or tubular heat exchangers can be used for relatively thin (low-viscosity) liquids. For viscous liquids, a scraped surface heat exchanger can be used to promote faster heat transfer and minimize surface fouling problems. These different types of heat exchangers are illustrated in Figure 6.1. Solid or semi-solid foods are first filled and sealed in containers, which undergo a canning process at atmospheric pressure. A more detailed description of the equipment systems used for canning is given later in Section 6.4 on canning.

6.2.2 Sterilization

Sterilization implies the destruction of all viable microorganisms. The term is commonly used in reference to the food canning process because canned foods are shelf-stable and seem never to experience microbial spoilage. Yet these foods are far from being sterile in the medical sense of the word. The success of the canning process (referred to as 'thermal processing' in the food industry) does not lie in destroying all viable microorganisms, but in the fact that, together with the food chemistry (pH) and microenvironment within the container (partial vacuum), hermetic packaging, and storage temperature less than that required for growth of thermophililic bacteria, the given thermal process prevents the growth of microorganisms of spoilage-causing and public health concern. In essence, it is a thermal process in which foods are exposed to a high enough temperature for a sufficiently long time to render them 'commercially' sterile.

Determining the time and temperature for a thermal sterilization process takes into account the thermal inactivation kinetics (heat resistance) of the target microorganism, as well as its sensitivity to oxygen, pH, and temperature. The absence of oxygen in the container prevents the growth of aerobic microorganisms and if the storage temperature is kept below 25 °C, highly heat-resistant spore-forming thermophilic bacteria pose little or no problem. From the public health perspective, the microorganism of greatest concern in low-acid foods with pH > 4.5 is *Clostridium botulinum*. This is a heat-resistant, anaerobic bacterium. If it survives processing, it can potentially grow and produce deadly botulism toxin in a canned food. *Clostridium botulinum* is a spore-forming bacteria and, like most spore-forming bacteria, cannot grow at pH< 4.5 (acid and acidified foods). The target microorganisms in acid or

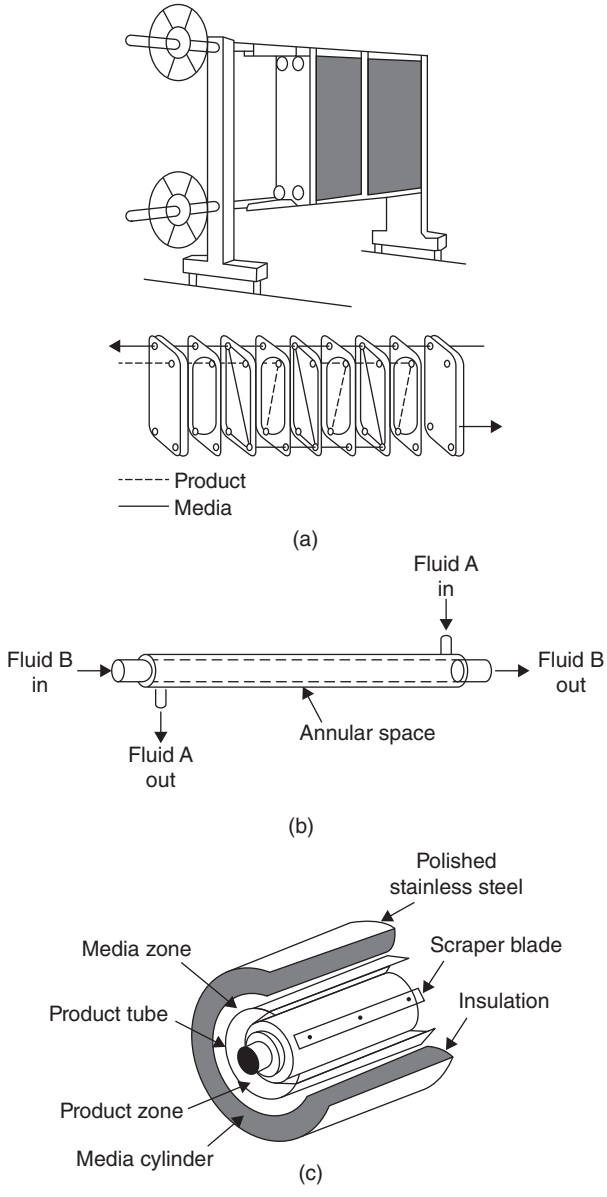

Figure 6.1 (a) Plate heat exchanger with product flow schematic, (b) schematic of tubular heat exchangers, and (c) cutaway section of the swept surface heat exchanger (source: Singh and Heldman, 2009. Reproduced with permission of Elsevier)

acidified foods consist of yeasts, molds, and vegetative microorganisms with relatively low heat resistance. These foods can be made shelf-stable when processed at pasteurization temperatures below the boiling point of water.

In contrast, spore-forming bacteria that can grow in low-acid foods are far more heat resistant and require temperatures well above the atmospheric boiling point of water for thermal inactivation. Therefore, low-acid foods must be processed with equipment systems that operate under pressure, such as pressurized retorts or autoclaves used in canning for in-container sterilization, or ultra-high-temperature (UHT) processing of liquid products in heat exchanger systems through which the product flows under pressure controlled by a back-pressure valve. A preferred alternative to heat exchanger systems for UHT processing is the use of steam injection or steam infusion. In these systems pressurized live steam of culinary quality is injected directly into the liquid product just prior to entering the hold tube. This causes the temperature of the resulting mixture of steam condensate and liquid product to rise instantaneously to the sterilizing temperature. Instantaneous cooling is achieved when the mixture exits the hold tube into a pressure-controlled flash chamber. Steam injection is accomplished with the use of a nozzle that injects steam directly into the product. Steam infusion is accomplished when the product enters a pressurized steam chamber prior to entering the hold tube, as shown in Figure 6.2.

The UHT process is used to produce long shelf-life refrigerated low-acid liquid products, such as UHT milk or cream. Shelf-stable low-acid liquid products require aseptic processing, in which the cool UHT sterilized product is filled into sterile containers within a sterile atmosphere to avoid post-process contamination from air-borne microorganisms. A more detailed description of aseptic processing of sterilized shelf-stable liquid products is given in the following section. Equipment systems used for in-container sterilization, such as pressurized retorts or autoclaves, are further described in Section 6.4 on canning.

6.3 Aseptic processing

Among the first commercially successful aseptic canning systems was the Dole Engineering system. It was designed to fill conventional steel cans aseptically and made use of superheated steam chambers to sterilize empty can bodies and covers as they were slowly conveyed to the filling chamber. The filling chamber was also maintained sterile by superheated steam under positive pressure and received cool sterile product from the heat exchangers in the product UHT sterilizing system. The entire system was sterilized prior to operation by passing superheated steam through the can tunnel, cover and closing chamber, and filling chamber for a prescribed start-up program of specified times and temperatures. The UHT sterilizing system

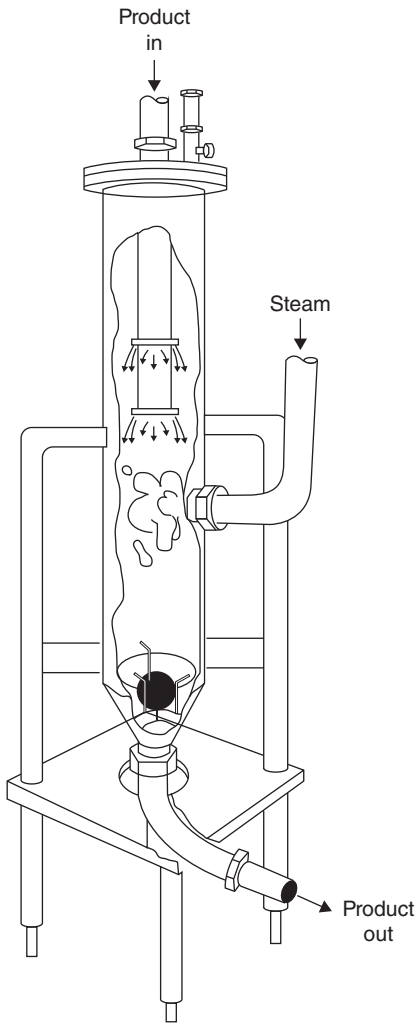

Figure 6.2 Schematic of steam infusion heat exchanger (source: Singh and Heldman, 2009. Reproduced with permission of Elsevier)

was pre-sterilized by passing pressurized hot water through the cooling heat exchanger (with the coolant turned off), product filling line, and filler heads. This start-up procedure had to be repeated every time a compromise in sterility occurred at any system component. Obviously, careful monitoring and control by skillful and highly trained operators is a must for such an intricately orchestrated system.

Regulatory approval for the use of chemical sterilizing agents, such as hydrogen peroxide, to sterilize the surfaces of various paper, plastic, and laminated packaging materials opened the door to a wide array of

commercially available aseptic filling systems to produce shelf-stable liquid foods in a variety of gable-topped, brick-packed, and other novel package configurations. Filling machines designed for these packaging systems are usually based on the use of form-fill-seal operations. Packaging material is fed from either pre-cut blanks or directly from roll stock, passed through a chemical sterilizing bath or spray treatment, are formed into the final package shape while being filled with cool sterile product from the UHT product sterilizing system, and then sealed and discharged, all within a controlled aseptic environment.

Another important commercial application of aseptic processing technology is in the storage and handling of large bulk quantities of sterilized food ingredients, such as tomato paste, fruit juice concentrates and purees, and other liquid food concentrates that need to be purchased by food processors or institutional end users for use as ingredients in further processed prepared foods. The containers for such applications can range in size from the classic 55-gallon steel drum to railroad tank cars or stationary storage tanks. Specially designed aseptic transfer valves and related handling systems make it possible to transfer sterile products from one such container to another without compromising sterility. The filling of pre-sterilized fermentation reactors with in-line UHT-sterilized liquid substrates is a long-standing classic example of aseptic filling of large bulk containers.

6.4 Canning

Food canning is most often referred to as thermal processing within the food industry. Thermal processing consists of heating food containers in pressurized retorts (autoclaves or 'pressure cookers') at specified temperatures for prescribed lengths of time. These process times are calculated on the basis of achieving sufficient bacterial inactivation (lethality) in each container to assure food safety to the consuming public and to ensure that the probability of spoilage will be less than some minimum. Associated with each thermal process is always some degradation of heat-sensitive vitamins and other quality factors that is undesirable. Because of these quality and safety factors, great care is taken in the calculation of process times and in the control of time and temperature during processing to avoid either under- or overprocessing. The methods by which these process times and temperatures are calculated and the scientific principles upon which they are based can be found elsewhere (Teixeira, 2007a, 2007b; Lopez, 1987; Stumbo, 1965; Ball and Olson, 1957). This section describes some of the commercial retort equipment systems that are used in the food canning industry to accomplish thermal processing efficiently on a production scale. Just as with most industrial processing operations, both batch and continuous systems are available. As the name implies, batch systems are made up of individual batch retorts that operate

intermittently. Scheduling of the retorts is skillfully staggered so that workers move from retort to retort, manually unloading and reloading each retort as its scheduled process cycle comes to an end. In continuous systems, cans are automatically fed into and out of retort systems that operate continuously over one or more working shifts.

6.4.1 Batch retort systems

Batch retorts are large pressure vessels in which baskets or crates of food containers are sterilized with saturated steam or water under pressure. These retorts can be either vertical or horizontal in configuration. The vertical still-cook batch retort is perhaps the grandfather of all batch retorts. Hardly any food science pilot plant or laboratory is complete without one. A typical production vertical unit will measure nearly 4 ft in diameter by 8 or 9 ft in height. Cans are loaded in baskets that are handled by chain hoist for lifting and lowering into the vertical retort. In the case of horizontal retorts, cans are loaded into wheeled carts that roll on rails into and out of the retorts. Most retorts are designed to hold either three or four crates or carts, with a total capacity of more than 2000 pint-size cans or 400 gallon-size cans. Although the basic design of these retorts has changed little since the turn of the twentieth century, they are still quite popular and can be found operating in many food canneries today. Part of the reason for this continued popularity is the simplicity of their design and operation and their versatility to accommodate virtually all can sizes and shapes.

Although the unloading and reloading operations are labour intensive, a well-managed cook room can operate with surprising efficiency. The cook room is the room or area within a food canning plant in which the retorts are located. Some cook rooms are known to have more than 100 vertical still cook retorts operating at full production. Although each retort is a batch cook operation, the cook room as a whole operates as a continuous production line, in that filled and sealed unsterilized cans enter the cook room continuously from the filling line operations and fully processed sterilized cans leave the cook room continuously. Within the cook room itself, teams of factory workers move from retort to retort to carry out loading and unloading operations, while retort operators are responsible for a given number of retorts. These operators carefully monitor the operation of each retort to make sure that the scheduled process is delivered for each batch.

For convection-heating products that benefit from mechanical agitation during processing, agitating batch retorts are available. A modern-day horizontal batch retort is shown in Figure 6.3, with a battery of several such retorts making up a large cook-room operation, shown in Figure 6.4. Batch retorts designed for flexible or semi-rigid retortable packaging systems require overriding air pressure to protect packages from bursting during processing. These operate with water spray, water cascade, or steam–air mixtures with

Figure 6.3 Modern-day horizontal batch retort (source: Photo courtesy of JBT FoodTech, formerly FMC FoodTech, Madera, CA). *See plate section for colour version*

Figure 6.4 Batch retort system showing battery of retorts in a large cook room operation (source: Photo courtesy of JBT FoodTech, formerly FMC FoodTech, Madera, CA). *See plate section for colour version*

overriding air pressure, and are capable of delivering end-over-end agitation when desired.

6.4.2 Continuous retort systems

Continuous retort operations require some means by which filled, sealed containers are automatically and continuously moved from atmospheric conditions into a pressurized steam environment, held or conveyed through that environment for the specified process time, and then returned to atmospheric conditions for further down-stream handling operations. The best-known

commercially available systems that accomplish these requirements are the crateless retort, the continuous rotary cooker, and the hydrostatic sterilizer.

Crateless retorts A crateless retort system is, in a sense, an automatic cook room in that the system is made up of a series of individual retorts, each operating in a batch mode, with loading, unloading, and process scheduling operations all carried out automatically without the use of crates. When ready to load, the top hatch opens automatically and cans fed from an incoming conveyor literally 'fall' into the retort, which is filled with hot water to cushion the fall. Once fully charged, the hatch is closed and steam entering from the top displaces the cushion water out of the bottom. When the cushion water has been fully displaced, all valves are closed and processing begins. At the end of the process time, the retort is refilled with warm water and the bottom hatch, which lies beneath the water level in the discharge cooling canal, is opened to let the cans fall gently on to the moving discharge conveyor in the cooling canal. After all cans are discharged, the bottom hatch is reclosed and the retort is ready to begin a new cycle. A commercial system of crateless retorts would consist of several such retorts in a row sharing a common in-feed and discharge conveyor system to achieve continuous operation of any chosen capacity.

Continuous rotary cookers The continuous rotary pressure sterilizer or 'cooker' is a horizontal rotating retort through which the cans are conveyed while they rotate about their own axis through a spiral path and rotating reel mechanism, as illustrated in the cutaway view and schematic in Figure 6.5. Residence time through the sterilizer is controlled by the rotating speed of the reel, which can be adjusted to achieve the required process time. This, in turn, sets the line speed for the entire system. Cans are transferred from an incoming can conveyor through a synchronized feeding device to a rotary transfer valve, which indexes the cans into the sterilizer while preventing the escape of steam and loss of pressure. Once cans have entered the sterilizer, they travel in the annular space between the reel and the shell. They are held between spines on the reel and a helical or spiral track is welded to the shell. In this way the cans are carried by the reel around the inner circumference of the shell, imparting a rotation about their own axes, while the spiral track in the shell directs the cans forward along the length of the sterilizer by one can length for each revolution of the reel. At the end of the sterilizer, cans are ejected from the reel into another rotary valve and into the next shell for either additional cooking or cooling.

Most common systems require at least three shells in series to accomplish controlled cooling through both a pressure cool shell and an atmospheric cool shell following the cooker or sterilizer. For cold-fill products that require

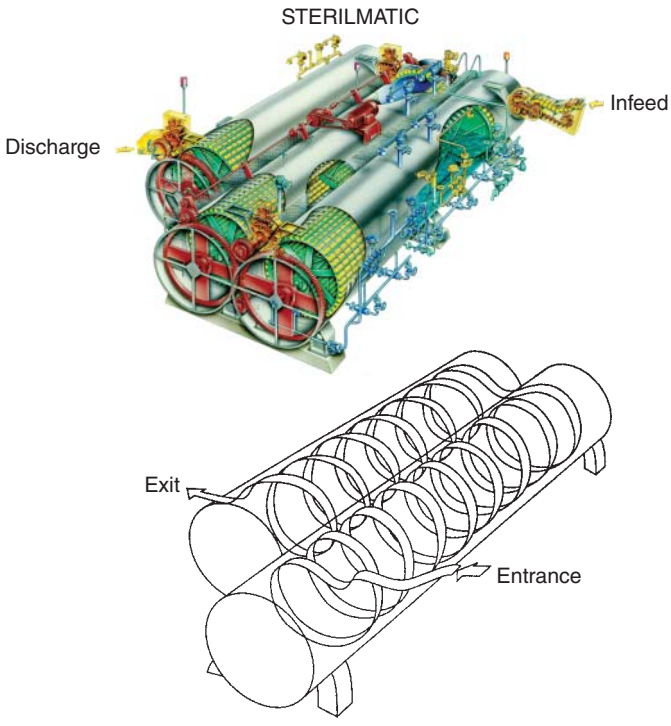

Figure 6.5 Cutaway view of continuous horizontal rotary cooker/sterilizer (top) and schematic of helical path of travel through each rotary cooker (bottom) (source: Photo courtesy of JBT FoodTech, formerly FMC FoodTech, Madera, CA)

controlled pre-heating, as many as five shells may be required in order to deliver an atmospheric pre-heat, pressure pre-heat, pressure cook, pressure cool, and atmospheric cool. By nature of its design and principle of operation, a continuous rotary sterilizer system is manufactured to accommodate a specific can size and cannot easily be adapted to other sizes. For this reason it is not uncommon to see several systems (filling lines) in operation in one food canning plant, each system dedicated to a different can size.

Hydrostatic sterilizers Hydrostatic sterilizer systems are so named because steam pressure is controlled hydrostatically by the height of a leg of water. Because of the height of water leg required (over 10 meters), these sterilizers are usually installed outdoors adjacent to a canning plant. They are self-contained structures with the external appearance of a rectangular tower, as shown in Figure 6.6. They are basically made up of four chambers: a hydrostatic 'bring-up' leg, a sterilizing steam dome, a hydrostatic 'bring-down' leg, and a cooling section.

Figure 6.6 Exterior view of a continuous hydrostatic sterilizer (source: Photo courtesy of JBT FoodTech, formerly FMC FoodTech, Madera, CA). *See plate section for colour version*

The principle of operation for a hydrostatic sterilizer can be explained with reference to the schematic diagram in Figure 6.7. Containers are conveyed through the sterilizer on carriers connected to a continuous chain link mechanism that provides positive line speed control and, thus, residence-time control to achieve a specified process time in the steam dome. Carriers are loaded automatically from incoming can conveyors and travel to the top of the sterilizer, where they enter the bring-up water leg. They travel downward through this leg as they encounter progressively hotter water. As they enter the bottom of the steam dome, the water temperature will be in equilibrium with the steam temperature at the water–steam interface. In the steam dome, the cans are exposed to the specified process or 'retort' temperature for the prescribed process time, which is controlled by the carrier line speed. When cans exit the steam dome, they again pass through the water seal interface at the bottom and travel upward through the cool-down leg as they encounter progressively cooler water until they exit at the top. Cans are then sprayed with cooling water as the carriers travel down the outside of the sterilizer on their return to the discharge conveyor station.

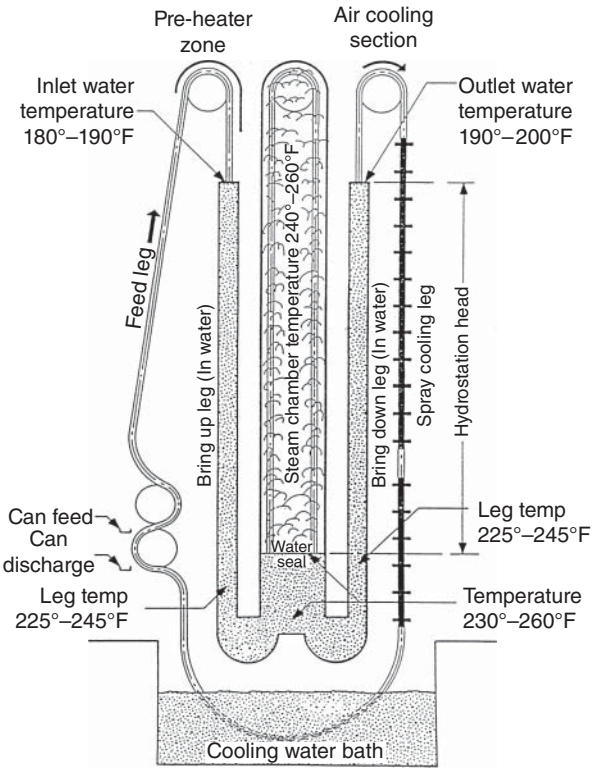

Figure 6.7 Schematic diagram of a hydrostatic sterilizer, illustrating the principle of operation (source: Photo courtesy of JBT FoodTech, formerly FMC FoodTech, Madera, CA)

6.5 Blanching

Blanching is a relatively mild form of heat treatment consisting of a few minutes of exposure to boiling water or atmospheric steam, and is not a food preservation process. It is normally applied to fresh foods (fruits and vegetables) prior to further processing. In food processing, blanching is usually the last step in the sequence of raw material preparation (washing, sorting, cutting, chopping, dicing, slicing, etc.). It is often needed to release entrapped air and inactivate enzymes prior to down-stream preservation processes such as drying, freezing, and canning. In the food process industry, blanching is normally accomplished by having the prepared raw food material travel on a conveyor through an atmospheric steam tunnel or hot water bath.

In the context of food preparation in cooking, blanching is an alternative term for par boiling. It is a heating process wherein the food is plunged into boiling water for just a few minutes and then quickly immersed in cold water (shocked) to immediately halt the cooking process. The meaning of blanching is 'to whiten', but this is not always the purpose of blanching in cooking. Food is often blanched to soften it, or to partly or fully cook it, or to remove a strong undesirable taste (for example, of bacon, cabbage, or onions).

References

Ball, C.O. and Olson, F.C.W. (1957) *Sterilization in Food Technology*. McGraw-Hill, New York, USA.

Lopez, A. (1987) *A Complete Course in Canning*, Book 1, *Basic Information on Canning*, 12th edn. CTI Publications, Inc., Baltimore, Maryland, USA.

Singh, R.P. and Heldman, D.R. (2009) *Introduction to Food Engineering*, 4th edn. Elsevier Publishing, London, England.

Stumbo, C.R. (1965) *Thermobacteriology in Food Processing*. Academic Press, New York, USA.

Teixeira, A.A. (2007a) Thermal processing of canned foods. Chapter 11 in: Heldman D.R. and Lund, D.B. (eds), *Food Engineering Handbook*, 2nd edn. CRC Press, Taylor & Francis Group, Boca Raton, Florida, USA, pp. 592–659.

Teixeira, A.A. (2007b) Thermal processing for food sterilization and preservation. Chapter 15 in: Kutz, M. (ed.), *Handbook of Farm, Dairy, and Food Machinery*. William Andrew Publishing, Springer Press, Norwich, New York, USA (now Elsevier Publishing, London, England), pp. 415–448.

7
Extraction Processes

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7.1 Introduction

Extraction is a separation technique employed in several food industries. It is essentially the process of moving one or more compounds of interest (analytes) from one phase or their original location (usually referred as the sample or matrix) to another phase or physically separated location where further processing and analysis occurs (Bicking, 2000). The extraction can be from solid to liquid, liquid to liquid and so on. The use of a convenient type of extraction not only influences the accuracy of the results but also determines the total analysis time and in this way affects the sample and the analysis. The extraction process can be categorized into two classes – conventional and advanced techniques.

7.2 Conventional extraction

The process of separation of compounds is carried out by vapour phase enhancement, partition with solvents involving phase separation and also the combination with vapour phase enhancement based on distillation.

7.2.1 Separation of steam volatiles

This is possibly the oldest extraction method used for the separation of essential oils from plant materials. It is based on the partial pressures in a mixture and the enhancement of the vapour phase of the components in a mixture by

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increasing the temperature of the mixture. A mixture is heated in water; the resulting steam is composed of components based on the relative vapour pressure of the components at the steam temperature. When the steam condenses, the organic compounds separate out as a water-insoluble phase. Heat energy is used to convert the organic compounds in the mixture or solid matrix to the vapour phase. The plant material is subjected to a temperature of 100 °C by steam at atmospheric pressure. The limitation of this method is the alteration of the chemical constituents of the essential oils due to the thermolabile nature of the natural components. This process is limited to volatile organic compounds as they are insoluble in water.

7.2.2 Solvent extraction

A compound soluble in another compound that is chemically similar is the principle behind the process of solvent extraction. It is used to extract a wide range of compounds from the plant materials using different organic solvents. The selection of the solvent is based on the nature (polar or nonpolar) of the compound to be extracted.

This extraction apparatus was developed by the German chemist Franz von Soxhlet in 1879. In the conventional Soxhlet system, the plant material is placed in a thimble-holder and is filled with condensed fresh solvent from a distillation flask. When the liquid reaches the overflow level, a siphon aspirates the solution of the thimble-holder and unloads it back into the distillation flask, carrying extracted solutes into the bulk liquid. In the solvent flask, the solute is separated from the solvent using the process of distillation. The solute is left in the flask and fresh solvent passes back into the plant solid bed. The operation is repeated until complete extraction is achieved. Some of the disadvantages of the Soxhlet system is that (a) the different solvents should be used to extract the different nutraceuticals compounds, (b) it requires a large amount of solvent, and (c) it takes a longer time for complete extraction.

Conventional extraction techniques require a longer time and larger amounts of solvent for the extraction. The possibility of thermal degradation of the target compound cannot be ignored as the extraction usually occurs at the boiling point of the solvent for a long time. To overcome these problems, some advanced techniques have been designed.

7.3 Advanced extraction processes

7.3.1 Ultrasound assisted extraction

Sound waves that have frequencies higher than 20 kHz can travel in a material, and involve expansion and compression cycles during travel in the

medium. These expansions and compressions create a bubble and collapse at a solid surface, which has a greater impact on the yield and extraction kinetics. This increases the penetrating power of the solvent into cellular materials and improves mass transfer. Ultrasound in the extraction process can also disrupt biological cell walls and facilitate the release of the inner contents. Compared to other extraction techniques like microwave assisted extraction, ultrasound assisted extraction (UAE) is cheaper and its operation is easier (Wang and Weller, 2006).

7.3.2 Microwave assisted extraction

Microwaves are electromagnetic radiations with a frequency from 0.3 to 300 GHz. Microwaves can penetrate biomaterials and interact with polar molecules such as water in biomaterials to generate heat. Microwave assisted extraction (MAE) offers a rapid delivery of energy to a total volume of solvent and solid plant matrix. It heats the solvent and solid matrix efficiently and homogeneously. Because water within the plant matrix absorbs microwave energy, cell disruption is promoted by internal super heating, which facilitates the desorption of chemicals from the matrix and improves the recovery of nutraceuticals. Like UAE, MAE also reduces the time of extraction, solvent usage and increases the extraction yield. The disadvantage of this technique is that an additional filtration or centrifugation step is necessary to remove the solid residues during MAE. Generation of microwaves is, however, a costly processing step.

7.3.3 High pressure extraction

High pressure extraction (HPE) is the process of extraction of bioactive components using high pressure solvents like supercritical fluids. Conventional processes are limited by numerous disadvantages: decomposition of thermally labile bioactive compounds because of high temperature and long duration of heat treatment, nonselective extraction of bioactive compounds and the need to reduce the toxicity of the solvents. Even though the solvating power of supercritical fluids has been demonstrated (Hannay and Hogarth, 1879) more than a century back, supercritical fluid techniques are widely used in analytical as well as on an industrial scale since the commercial success of decaffeination of coffee (Zosel, 1978). A fluid heated to above the critical temperature and compressed to above the critical pressure is known as a supercritical fluid. It has the unique ability to diffuse through a solid matrix like a gas and dissolves materials like a liquid. Additionally, it can readily change its density upon minor changes in temperature or pressure. Carbon dioxide and water are the most commonly used supercritical fluids. Carbon dioxide has a low critical temperature and moderate pressure of 31.1 °C and

73 bar, respectively. Water has a critical temperature of 647 K and a critical pressure of 220 bar due to its high polarity.

Supercritical fluids (SCFs) such as water and carbon dioxide are substances that are compatible with the Earth's environment. However, several other supercritical fluids can be used, but the final choice would depend on the specific application and additional factors such as safety, flammability, phase behaviour and solubility at the operating conditions and the cost of the fluid. The basic principle of SCF extraction is that the solubility of a given compound (solute) in a solvent varies with both temperature and pressure. At the ambient conditions (25 °C and 1 bar), the solubility of a solute in a gas is usually directly related to the vapour pressure of the solute and is generally negligible. In a SCF, however, the solute solubility is up by 10 orders of magnitude greater than those predicted by the ideal gas law behaviour.

The critical point is positioned at the end of the gas–liquid equilibrium curve where the supercritical fluid region is also placed. It can be shown that by using a combination of isobaric changes in temperature with isothermal changes in pressure, it is possible to convert a pure component from a liquid to a gas (and vice versa) via the supercritical region without incurring a phase transition.

Physical properties of supercritical fluids Above the critical temperature of a compound, the pure gaseous component cannot be liquefied regardless of the pressure applied. In the supercritical environment only one phase exists. The fluid, as it is termed, is neither a gas nor a liquid and is best described as an intermediate of the two extremes. The physical properties of SCFs are in between those of a gaseous and liquid state. Different physical properties like density and viscosity of SCFs are available (Martinez, 2008). Thermal conductivities are relatively high in the supercritical state and have very large values near the critical point (CP) because, in principle, the heat capacity of a fluid tends to infinity at the CP. Interfacial tension is close to zero in the critical region. In general, the physical properties in the critical region enhance the mass and heat transfer processes.

The basic principle of supercritical fluid extraction (SFE) is that when the feed material is contacted with a supercritical fluid (SCF), then the volatile substances will partition into the supercritical phase. After the dissolution of the soluble material, the supercritical fluid containing the dissolved substances is removed from the feed material. The extracted component is then completely separated from the SCF by means of a temperature and/or pressure change. The SCF may be recompressed to the extraction conditions and recycled.

In the last decade, new trends have emerged in the food industries. These trends include an enhanced concern for the quality and safety of food products, increased preference for natural products over synthetic ones and broadened regulations related to nutritional and toxicity levels

of active ingredients. Consumers are more educated and health conscious than ever before and are demanding higher quality products for consumption. Hence, alternative extraction methodologies complying with both consumer preference and regulatory controls, as well as cost effectiveness are becoming popular. One such major technology that has emerged over the last two decades, as an alternative to the traditional solvent extraction of natural products, is the SFE technology. Though the solvent properties of supercritical fluids (SCF) were recognized over 100 years ago, their commercial applications are relatively recent, possibly due to the requirement for sophisticated and expensive high pressure equipment and technology. SFE is a relatively new separation unit operational to solve some of the problems of conventional separation techniques, like distillation and solvent extraction. SFE uses a clean, safe, inexpensive, nonflammable, nontoxic, environmental friendly and nonpolluting solvent like carbon dioxide and the energy costs of this novel extraction technique are lower than those for the traditional solvent extraction methods. It also displays a wide spectrum of solvation power as its density is strongly dependent on both temperature and pressure–temperature swings. This unique feature facilitates solute recovery, the 'fine-tuning' of solution power and the fractionation of mixed solutes. SFE is thus increasingly gaining importance over the conventional techniques for extraction of natural products.

Solvents used as SCFs The choice of the SFE solvent is similar to the regular extraction processes and depends on what type of compound is to be extracted. The principle considerations are the good solvating property, inert to the product, easy separation from the product and cheap. Critical data on some supercritical solvents are available in different handbooks.

Carbon dioxide is the most commonly used SCF primarily due to its low critical parameters (31.1 °C, 73.8 bar), low cost and nontoxicity. However, several other SCFs have been used in both commercial and developmental processes. Organic solvents are usually explosive so a SFE unit working with them should be explosion proof; this fact makes the investment more expensive. Organic solvents are mainly used in the petroleum chemistry. CFCs (chlorofluorocarbons) are very good solvents in SFE due to their high density, but the industrial use of chlorofluoro hydrocarbons are restricted because of their effect on the ozonosphere. Therefore, CO_2 is the most widely used fluid in SFE. Beside CO_2, water is the other increasingly applied solvent. One of the unique properties of water is that, above its critical point (374 °C, 218 atm), it becomes an excellent solvent for organic compounds and a very poor solvent for inorganic salts. This property gives the chance for using the same solvent to extract the inorganic and the organic components, respectively.

Solubility of SCFs with analogy to liquids Supercritical fluid extraction (SFE) is a separation technique that exploits the solvent power of supercritical fluids

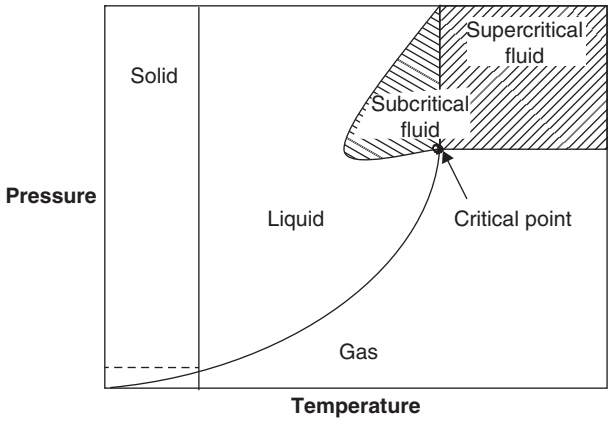

Figure 7.1 A generalized phase diagram for a pure liquid

(SCFs) at temperatures and pressures near and above the critical point of the fluids. A supercritical fluid is the state of a substance above the critical temperature (T_c) and critical pressure (P_c).

Figure 7.1 presents a generalized pressure–temperature (P–T) phase diagram for a pure fluid, in which the regions solid (s), liquid (l) and gas (g) are demarcated. The lines indicted the coexistence of two phases. T is the triple point, where all three phases exist, and C is the critical point, where the properties of the liquid and vapour become identical. The supercritical region is shown as the hatched region, which is of importance to industries.

If a gas like carbon dioxide or ethylene is compressed at temperatures above its critical point, it is not possible to liquefy the gas no matter however high the pressure is applied. The critical temperature (T_c) is defined as the temperature just above where it is impossible to liquefy the gas. The critical pressure (P_c) may be defined as the pressure required to liquefy the vapour at the critical temperature (T_c). In brief, the critical pressure is the highest vapour of the liquid of that substance. The reduced pressure (P_r) is the ratio of the pressure of the fluid to the critical pressure, the reduced temperature (T_r) is the ratio of the temperature to the critical temperature and the reduced density (ρ_r) is the ratio of the density to the critical density.

The solvent power of supercritical fluids can be understood by analogy with the solubility of substances in liquids/solvents. This can be better explained by considering the density pressure isotherms for a typical gas solvent. The diagram is plotted in terms of the reduced variables like the reduced temperature (T_r), reduced pressure (P_r) and reduced density (ρ_r).

The solvent power is attributed to the intermolecular forces resulting from the close packing of solvent molecules around the solute molecules, which is generally related to the density of the liquid. At temperatures just above the critical temperature ($1.06T_r$), the densities close to liquid

densities can be achieved. Therefore the supercritical fluids having densities of the liquid can solubilize materials. SCF extraction is generally carried out at temperatures less than $T_r = 2$; as T_r increases, it needs a very high pressure to keep the density of the fluids reasonably close to that of liquids. Further, the density of the SCFs can be varied by varying the pressure and temperature. Thus, it can affect the variation of solvencies by changing the system pressure and temperature. This high sensitivity of SCFs to the system pressure and temperature makes supercritical fluid extraction a potential economic process. Thus, the region of interest is the SCF region, shown by the hatched area in Figure 7.1.

At constant pressure, the solvent power of the supercritical fluid increases as the temperature is reduced, whereas the solubility increases with the pressure at constant temperature. With increasing temperature, relatively higher pressures are needed to have a better loading of the solute in the solvent. At supercritical temperatures and subcritical pressures, the solubility is low. As the supercritical phase is isothermally compressed (say 0.5 Pr), the dissolved material would be precipitated. By suitable variations of process conditions like temperature and pressure, SCFs can be used for extraction of specific components that can be separated easily from the fluid, allowing it to the recovered and reused.

A brief theory of SCFs At low pressures the solubility of a solid in a gas is very small. At equilibrium, the partial pressure of the solute in the gas phase equals the vapour pressure of the pure solid. The assumption of ideal gas phase behaviour of the solubility is given by

$$y_S = \frac{P_2^s}{P} \tag{7.1}$$

neglecting the solubility of the gas in the solid. As the temperature and pressure approach and exceed the critical temperature and pressure of the solvent gas, the situation changes. As indicated earlier, the gas becomes more liquid like in its solvent properties and the solubility of the solid may increase markedly. A solubility enhancement factor (E)

$$y_2 = E\frac{P_2^s}{P} \tag{7.2}$$

represents the ratio of the actual solubility to the ideal gas solubility. Here, P_2^s is the vapour pressure of the solid and P is the total pressure of the system. This enhancement factor of solubility provides the basis for the supercritical extraction processes. Enhancement factors of the order 10^4 are common and systems having enhancement factors of 10^{10} are known.

Enhancement factors can be calculated thermodynamically using the equation of state. An example using the virial equation of state is illustrated below:

$$\frac{PV}{RT} = 1 + \frac{B}{V} + \frac{C}{V^2} + \frac{D}{V^3} + \cdots \tag{7.3}$$

Here B, C, D, etc., are the virial coefficients. The terms P, V, R and T are the terms of the ideal gas law. Rowlison and Richardson (1959) derived the relationship

$$\ln \ E = \frac{V_2^S - 2B_{12}}{V} \tag{7.4}$$

where B_{12} is the cross-virial coefficient and represents the interaction between the molecules of gas solvent 1 and solute 2. The greater the interaction, the more is the negative value of B_{12} and the greater the enhancement factor and hence the solubility. Here, V is the molar volume and $V_2{}^s$ is the molar volume of the solid or solute.

The second virial coefficient (B) for the pure gases can be estimated from the corresponding status relationship. Pitzer and Curl (1957) gave the following most successful relationship:

$$\frac{BP_c}{RT_c} = f_0(T_r) + \omega f_1(T_r) \tag{7.5}$$

where ω is the eccentric factor, a measure of the complexity of molecules with respect to both its nonsphericity and its polarity. The terms P_c, T_c and T_r are critical pressure, critical temperature and reduced temperature, respectively. The tabulated values of these fractions are given by Reid, Prausnitz and Sherwood (1977):

$$T_{r12} = (T_{c1}\, T_{c2})^{1/2}(1 - k_{12}) \tag{7.6}$$

$$V_{c12} = \left(V_{c1}^{1/3} + V_{c2}^{1/3} \right)^{3/2} \tag{7.7}$$

$$P_{c12} = \frac{(Z_{c12} R T_{c12})}{V_{c12}} \tag{7.8}$$

$$Z_{c12} = \frac{(Z_{c1} + Z_{c2})}{2} \tag{7.9}$$

$$\omega_{12} = \frac{\omega_1 + \omega_2}{2} \tag{7.10}$$

where Z_c is the critical compressibility factor, V_c is the critical volume and k_{12} is a binary constant. Lists of these constants are published for a variety of binary systems (Chueh and Prasunitz, 1967). These relations are probably reliable for molecules that do not differ markedly in size or chemical structure.

Equations (7.2) and (7.3) can be plotted for nonpolar gases. This shows that B_1, and hence B_{12}, is negative for most values of T_r, in which case, from Equation (7.3), E is positive.

Some general conclusions can be drawn from above. These are:

1. For a gas of given T_c, reducing the extraction T will decrease V and make B_{12} more negative and hence will increase E. However, reducing T will also reduce the solute vapour pressure P_2^s. As a result, the solubility of the solute will also reduce the vapour pressure P_2^s. Therefore the solubility of the solute is expected to have a maximum value at an optimum temperature, representing a compromise between the two effects.
2. For a given extraction temperature, gases of high T_c should have large negative values of B_{12} and hence are better solvents than those of low T_c.
3. An increase in the extraction pressure (P) will decrease the molar volume of the solute (V_s) and hence there is an increase in E and the solubility. However, if $V_2^S - B_{12}$ and P_2^S are small, for example if the temperature is too high or the solute has a too low volatility, excessive high pressure will be required to achieve adequate solubility.

In general, these conclusions are satisfactory as a rough guide as certain approximations are taken in deriving the equations. The virial equation has been truncated to the second virial coefficient. The solubility of the gas in the solute has been ignored. However, these general guidelines can safely be extended to the general systems. A more comprehensive thermodynamic approach will be dealt with in the subsequent sections.

Critical data The SCF exhibits physico-chemical properties intermediate to those of a gas and a liquid (Table 7.1), which enhances its role as a solvent. Its relatively high density gives a good solvent power, while its relatively low viscosity and diffusivity values provide appreciable penetrating power into the solute matrix. These properties give rise to higher rates of mass transfer of solutes into a supercritical fluid than into a liquid.

Some of the supercritical solvents that can be used in SFE are available. These solvents cover a wide range of critical temperature, molecular size and polarity. Many of these are abundant and low in cost. It is often convenient to use mixtures rather than pure compounds.

Experimental data illustrating general principles of enhanced solubility in supercritical fluids The solubility of *p*-iodochlorobenzene in ethylene at 298 K is close to the critical temperature of ethylene (282 K). At low pressures, the gas phase solubility is difficult to measure, while with an increasing pressure of ethylene above the critical temperature of 5 MPa, the gas phase concentrations of *p*-iodochlorobenzene increases by three orders of magnitude (Ewald, Jepson and Rowlison, 1953).

Table 7.1 Mass transfer equations (for extraction through fixed bed solids)

Relationship	Remarks	Reference
$Re = 90 - 4000$, $J_D = 2.06/\xi Re^{-0.575}$ $Sc = 0.6$	General	Treybal (1986)
$Re = 5000 - 10\,300$, $J_D = 20.4/\xi Re^{-0.815}$ $Sc = 0.6$	General	Treybal (1986)
$Re = 0.0016 - 55$, $J_D = 1.09/\xi\ Re^{-2/3}$ $Sc = 168 - 70\,600$	General	Treybal (1986)
$Re = 5 - 1500$, $J_D = 0.250/\xi\ Re^{-0.31}$ $Sc = 168 - 70\,600$	General	Treybal (1986)
$3 < Re < 3000$, $Sh = 2 + 1.1\ Sc^{1/3}\ Re^{-0.6}$	Applied for rapeseed oil extraction using SCF	Treybal (1986)

Symbols: J_D is the dimensionless mass transfer constant as defined in Equation (7.27) and ξ is the void volume.

It is important to carry out the extraction process near the critical temperature of the gas for phenanthrene in nitrogen, methane, carbon tetraflouride, carbon dioxide and ethylene at 313 K and 40 MPa (Eisenbeiss, 1964). Those having the critical temperatures well above the extraction temperature, that is nitrogen (126 K), methane (191 K) and carbon tetraflouride (222 K), did not extract phenathrene, whereas the gases with a critical temperature near the extraction temperature were ethylene (283 K), carbon dioxide (304 K) and ethane (305 K), which proved effective as solvents.

The solubility of naphthalene in supercritical ethylene as a function of temperature at different pressures has been studied (Tsekhanskaya, Iomter and Muskina, 1964). At moderate pressures, a rise in temperature causes a decrease in solubility of the solid component. At high pressures, on the other hand, a rise in temperature causes an increase in the solubility. This contradictory behaviour of solubility with an increase in temperature can be interpreted in terms of the density of the gas. A rise in temperature at constant pressure leads to a decrease in gas density while offering an exponential increase in the vapour pressure of the solid. As illustrated earlier, slightly above the critical temperature, gas density at moderate pressures is lowered by a temperature rise to such an extent that the concentration of the solute in the supercritical phase decreases considerably. At high pressures, the decrease in density caused by the temperature rise is so small that the increase in the vapour pressure of the solid leads to higher concentrations in the supercritical phase.

Classification of phase behaviour of binary systems Phase diagrams are of considerable interest since they give pressure and temperature ranges of

complete mutual solubility and partial solubility. The simplest of the systems is a binary system consisting of component 1, the supercritical solvent, and component 2, the solid to be dissolved. These phase diagrams give qualitative insight into the solubility relations for the solids or liquids in equilibrium with the supercritical solvent. It is helpful to recall the Gibbs phase rule ($F = C - P + 2$) in interpreting the phase diagrams, where F is the number of degrees of freedom, C is the number of chemical components and P is the number of phases in equilibrium. For a binary system, $C = 2$ so that the maximum number of degrees of freedom is three for a single-phase system. A three-dimensional diagram is therefore required for a complete presentation of the phase relationships. The most commonly used independent variables in this diagram are pressure (P), temperature (T) and composition (x). However, various two-dimensional projections of this three-dimensional system are available.

The solid–fluid equilibrium is classified principally into two groups (Young, 1978; Schneider, 1978) depending on the shape of their critical curves. A more complete discussion and explanation of these phase relationships is given elsewhere (Street, 1983; Rowlison, 1969).

More complex phase diagrams occur where insoluble liquids appear and where more components are present. The basic ideas are similar; however, a similar opportunity for supercritical extraction exists. Now, a discussion on the binary phase behaviour of liquid–fluid systems is included. These are broadly classified as four types of the simplest kind of binary system phase behaviour.

Many other types of phase behaviour exist for the possible extraction of liquids. Simple binary diagrams can illustrate the behaviour that can find possible applications in different extraction processes.

Ternary systems For the simultaneous extraction of two components A and B with carbon dioxide, the phase diagram of the ternary system $A + B +$ carbon dioxide is of interest, especially when the concentrations of A and B are relatively high. The available ternary systems are for carbon dioxide + heptane + methanol, carbon dioxide + water + acetic acid, carbon dioxide + water + glycerides and carbon dioxide + butyl benzene + 2-methyl naphthalene.

Mass transfer phenomena Engineering design of the SFE process requires knowledge of equilibrium solubility used for the selectivity of components (i.e. thermodynamic constraints) as well as mass transfer rates. The mass transfer is influenced by a driving force and resistance in analogous fashion to the heat transfer process. The driving force is mainly due to the concentration gradient for the material being transported and the resistance is due to the medium through which material is being transferred and any interactions between that medium and the material.

In general, the mass transfer rate is equal to the driving force/resistance:

$$N_A = kA\Delta c \tag{7.11}$$

where N_A is the mass transfer rate (kg/s), A is the interfacial mass transfer area (s m^2), Δc is the concentration difference (kg/m^3 or kg/kg) and k is the mass transfer coefficient (m/s or kg/m^2 s). Equation (7.11) may also be written in terms of the diffusion coefficient:

$$N_A = -D_A \frac{\partial c_A}{\partial x} \tag{7.12}$$

where D_A is the diffusion coefficient and $\partial c_A/\partial x$ is the concentration gradient. Equation (7.12) is called Fick's law of diffusion. D is the characteristic of the component A and its environment (temperature, pressure, concentration and nature of other components in the material).

For the purpose mass transfer, it is assumed that the supercritical solvent flows through a fixed bed extracting the soluble components from the bed of solid material. Mass transfer phenomena in such a case occur in the following steps:

(a) external diffusion of solvent in the boundary layer,
(b) transfer of solvent into pores,
(c) solute–solvent interaction and solubilization of the solute,
(d) diffusion of solute into the pores, and
(e) external diffusion of the solute.

In most cases, the first three factors are not associated with the system design. These factors can also be improved by solvent viscosity and temperature. If the mass transfer rate is limited by the rate of diffusion of the solute into the pores, then the process is called 'internal mass transfer controlled'. If the rate of diffusion of the solute in the external gas phase controls the mass transfer, then the pressure is called 'external mass transfer controlled'.

Steady-state mass transfer When the concentration gradient remains unchanged with the passage of time, the rate of diffusion is also constant. Then Equation (7.12) for the flat slab of thickness x becomes

$$N_A = D\frac{\Delta c}{\Delta x} \tag{7.13}$$

For other shapes,

$$W = N_A S_{av} = D\,\frac{\Delta C}{\Delta x} \tag{7.14}$$

where $S_{av} = 2\pi(a_2 - a_1)/\ln(a_2 - a_1)$ for a solid cylinder and $S_{av} = 4\,\pi a_1 a_2$ for spheres, where a_1 and a_2 are the internal and external radii for spheres.

Mass transfer in a steady-state manner occurs when the solute concentration in the material is sufficiently high or during the initial period of solute extraction. Pressure, temperature of extraction and particle size of the material play significant roles in improving diffusion in this category of mass transfer phenomena.

Unsteady-state mass transfer Fick's second law of diffusion can also be used to solve unsteady-state mass transfer problems with appropriate boundary conditions:

$$\frac{\partial C}{\partial t} = \left[\frac{1}{x^{n-1}}\right] \frac{\partial \left[x^{n-1} D \frac{\partial C}{\partial x}\right]}{\partial x} \tag{7.15}$$

where $n = 1$ for the infinite slab, $n = 2$ for the infinite cylinder and $n = 3$ for the sphere. The equation describes how the concentration changes with time t and position x in the material. The method of solving the above equation depends upon the geometrical model chosen and upon the conditions at the boundaries.

Many assumptions are made to ease the solution to the above equation:

(a) the diffusion coefficient is constant,
(b) the mass transfer resistance in the gas phase is neglected, and
(c) the initial uniform concentration of the solute throughout the material.

The analytical solutions by way of infinite series for various geometrics are:
For the infinite slab,

$$\frac{C_i - C_t}{C_i - C_\infty} = \frac{8}{\pi^2} \sum_1^\infty \frac{1}{(2n-1)^2} \exp\left[-\left(\frac{(2n-1)}{2}\right)^2\right] \pi^2 F_o \tag{7.16}$$

For the infinite cylinder,

$$\frac{C_i - C_t}{C_t - C_\infty} = 4 \sum_1^\infty \frac{1}{B_n^2} \exp[-\mathrm{B}_n^2 F_o] \tag{7.17}$$

For the sphere,

$$\frac{C_i - C_t}{C_t - C_\infty} = \frac{6}{\pi^2} \sum_1^\infty n^{-2} \exp[-(n\pi)^2 F_o] \tag{7.18}$$

where, F_o is the Fourier number equal to Dt/L^2, where L is the characteristic length of the geometry of the material, B_n is the nth root of the equation, $T_0(B_n)$ is 0 (B_0 is the Bessel function of the first kind order of zero), C_i is the initial concentration of solute, C_0 is the surface solute concentration and C_t

is the average concentration of the solute at any time t. The solution is also available in the charts.

The unsteady state mass transfer can also be solved by penetration theory:

$$N_A = 2\ [C_{Ai} - C_{Ab}] \sqrt{\frac{D}{\pi t_e}} \tag{7.19}$$

where N_A is the average mass transfer rate of component A, C_{Ai} is the concentration of A at the interface, C_{Ab} is the concentration of A in the bulk and t_e is the time of exposure or extraction.

Equations (7.16), (7.17) and (7.18) are valid only when the mass transfer resistance in the gas phase is negligible. This is true when the Biot number, B_i (the ratio of external mass transfer conductance and internal mass transfer conductance), is greater than 100; when $B_i < 100$, Equations (7.16), (7.17) and (7.18) are to be modified correspondingly or appropriate lines in the charts are to be used.

When surface conditions (conditions in the gas phase) also influence the same mass transfer, the effects of these are reported in terms of dimensionless numbers, which are defined as follows:

$$\text{Reynolds number} : Re = \rho v \frac{L}{\mu} \tag{7.20}$$

$$\text{Sherwood number} : Sh = \frac{kL}{D} \tag{7.21}$$

$$\text{Schmidt number} : Sc = \frac{\mu}{\rho_D} \tag{7.22}$$

$$\text{Peclet number} : Pe = Re\ \ Sc \tag{7.23}$$

$$\text{Stanton number} : St = \frac{Sh}{Re\ \ Sc} \tag{7.24}$$

and

$$J_D = St\ \ Sc^{2/3} \tag{7.25}$$

where k is the mass transfer coefficient, μ is viscosity of the fluid, L is the characteristic length and ρ is density of the fluid.

The mass transfer analysis is usually reported in the form of

$$Sh = f(Re,\ \ Sc) \tag{7.26}$$

or

$$J_D = f\,(Re) \tag{7.27}$$

These are the equations available in the literature for various flow situations and geometries (Table 7.1). The relationships available are to be used with caution, as they may not be valid in supercritical conditions since buoyant effects play a role.

The importance of natural convection is measured by the ratio of buoyant to inertial forces. This is two orders of magnitude higher in a supercritical fluid (at a constant Reynold's number) than in normal liquid. Because of this factor, diffusivity without considering natural convection will lead to greater error. Mass transfer relationships are to be given in the form of Sherwood (Sh) and Grashof numbers (Debendetti and Reid, 1986).

$$Sh = f(Re, Sc, Gr) \tag{7.28}$$

$$Gr = \frac{\beta_T \Delta \mathrm{T} g L^3}{v^2} \tag{7.29}$$

Here β_T is thermal expansion coefficient, ΔT is temperature gradient, g is acceleration due to gravity, L is characteristic length, e.g. length in the direction of flow and v is kinematic viscosity. The large contribution of buoyant forces suggests that usual mass transfer correlations are unsuitable for design purposes when SCFs are involved.

The relationship between ξ and d_p is

$$a = 6(1 - \xi)/d_p \tag{7.30}$$

where a is the specific surface area or surface area per unit volume of bed, ξ is the void volume and d_p is the bulk density of the bed.

A crossover region is observed at higher pressures along with temperature for the mass transfer; at lower pressures, the mass transfer decreases with an increase in temperature, while at higher pressures it increases with an increase in temperature.

The effects of extraction pressure and temperature on extract yield and composition of the extract have been studied for several plant matters (Table 7.2). However, there are plenty of other process parameters like particle size, milling conditions, fluid velocity, etc., that influence the extraction process. Different approaches are available in the literature to obtain mathematical models that simulate supercritical fluid extraction from solid materials (Sovova, 2005, 1994; Martinez *et al.*, 2003; Cocero and Garcia, 2001; DeFranca and Meireles, 2000; Esquıvel, BernardoGil and King, 1999; Udaya Sankar and Manohar, 1994; Tan and Liou, 1989).

The selection of a suitable model to describe the extraction depends on:

(a) the physical meaning of parameters of a particular model,
(b) the simplicity or complex nature of the model, particularly the empirical ones,
(c) the fact that the model was used for a similar system by researchers, and
(d) the capability of the model to describe the trend of the overall extraction curve.

Table 7.2 Mass transfer data in the SFE system

System	SCF used	P(bar)	T(°C)	V(L/h)	K or D	Reference
Caffeine	Nitrous oxide	200	100	100	1.4×10^{-7} m/s	Brunner (1984)
Rapeseed oil	Carbon dioxide	205	51.5	200	4.0×10^{-4} m/s	Brunner (1984)
		350	40.8	200	3.4×10^{-4} m/s	Brunner (1984)
Rapeseed oil	Carbon dioxide	80–140	40, 60	200	0.08–0.425 g/cm^2 s	Gandhdhara Rao and Mukhopadhyay (1988)
Caffeine	Carbon dioxide	260	50.0	200	$D = 2.53 \times 10^{-6}$ cm^2/s	Udaya Sankar, Manohar and Chokkalingam Manohar (1986)
Pepper oil	Carbon dioxide	80–100	40, 60		$K = 4.85 \times 10^{-11}$ to 33.9×10^{-11}m/s	Udaya Sankar and Manohar (1988)
Sandalwood oil	Carbon dioxide	80	40		$K = 4.47 \times 10^{-7}$ to 4.9×10^{-6} m/s	Udaya Sankar and Manohar (1988)
Mango ginger extract	Carbon dioxide	100	40		0.799	Krishnamurthy (2012)
Mango ginger extract	Carbon dioxide	100	50		0.717	Krishnamurthy (2012)
Mango ginger extract	Carbon dioxide	100	60		0.546	Krishnamurthy (2012)
Mango ginger extract	Carbon dioxide	225	40		2.550	Krishnamurthy (2012)
Mango ginger extract	Carbon dioxide	225	50		1.650	Krishnamurthy (2012)
Mango ginger extract	Carbon dioxide	225	60		0.669	Krishnamurthy (2012)
Mango ginger extract	Carbon dioxide	350	40		1.690	Krishnamurthy (2012)
Mango ginger extract	Carbon dioxide	350	50		3.560	Krishnamurthy (2012)
Mango ginger extract	Carbon dioxide	350	60		18.50	Krishnamurthy (2012)

Almost every model in the literature treats the extract as a pure substance, although it may contain several compounds having a different chemical nature. This is more so when applied to natural plant materials used to describe the overall SFE process.

7.3.4 Applications of supercritical extraction of natural products

CO_2 offers the following advantages as a supercritical (SC) fluid:

- ✓ Being nontoxic and physiologically harmless, it has generally recognized as safe (GRAS) status.
- ✓ It is inflammable, noncorrosive and leaves behind no harmful residues after extraction. It does not cause any environmental pollution problem with $T_c = 31.1\,^{\circ}C$ and $P_c = 73.8$ bar. SC-CO_2 can be used at temperatures and pressures that are relatively safe, convenient and particularly appropriate for extraction of a wide range of compounds.
- ✓ Its near ambient critical temperature makes it suitable for thermolabile natural products, especially in food and pharmaceutical applications.
- ✓ It has good solvating power due to low viscosity and high diffusivity.
- ✓ Due to its low latent heat of vaporization, a low-energy input is required for the extract separation system, which renders the most natural odour and natural tasting extracts.
- ✓ The energy required for attaining the supercritical state in CO_2 is often less than the energy associated with the distillation of conventional organic solvents.
- ✓ CO_2 diffuses through condensed liquid phases (e.g. adsorbents and polymers) faster than typical solvents, which have larger molecular sizes (Perry and Green, 1999).
- ✓ In general, the extractability of compounds with SC-CO_2 depends on the presence of the individual functional groups in the compound, their molecular weights and polarity. SC-CO_2 is an excellent extraction medium for nonpolar species such as alkanes and terpenes and is reasonably good for moderately polar species like benzene derivatives, such as aldehydes and alcohols.
- ✓ As put forth by Brunner (2005), SC-CO_2 has the following qualities:
  - (i) It dissolves nonpolar or slightly polar compounds.
  - (ii) The solvent power for low molecular weight compounds is high and decreases with increasing molecular weight. Materials with a molecular weight greater than 500 Daltons have limited solubility in CO_2.
  - (iii) Free fatty acids and their glycerides exhibit low solubilities; low molecular weight nonpolar aliphatic hydrocarbons and small aromatic hydrocarbons are soluble.

(iv) Pigments are even less soluble.
(v) Water has a low solubility (<0.5% w/w) below 100 °C, and
(vi) Polar organics such as carboxylic acids, fruit acids, sugars, polysaccharides, proteins, phosphatides, glycosides and inorganic salts are relatively insoluble in dense CO_2.

The solvating power of SCF is highly dependent on its density. By controlling pressure and temperature, it is possible to control the density of the fluid allowing selective manipulation of the solvating power of the fluid. It is reported that solute solubility studies of individual components present in a mixture provide a means of determining SCF process conditions that will afford selective extraction or separation of the individual solutes (Mukhopadhyay, 2000). In the vicinity of the fluid critical point, physical properties of the fluid change dramatically with small changes in pressure and hence the solubility of the solutes changes dramatically; some authors have reported that by using this region, it is possible to separate mixed solutes (Raeissi and Peters, 2005).

7.3.5 Addition of co-solvents

The application of modifiers is probably the simplest yet most effective way to obtain a desired polarity of CO_2-based fluids. By selecting a modifier or just simply changing the molar ratio of a modifier, one can readily manipulate the solubility properties of the fluids. In general, the addition of a small amount of a liquid modifier can enhance significantly the extraction efficiency and consequently reduces the extraction time without significantly changing the density and compressibility of the original SCF solvent (Valcárcel and Tena, 1997).

The co-solvent mixed SC-CO_2 solvent is supercritical when the critical pressure and temperature of the mixture are above its mixture critical values for a particular composition, which are usually not very different from the critical values of the pure SC-CO_2.

In a binary mixture, when an SCF solvent and a co-solvent are employed beyond its binary mixture critical pressure to solubilize a liquid–solute, then the system is represented by a ternary diagram. In such cases, all three components are usually distributed both in the liquid and in the SC-CO_2 phases. The extent of solubilization of the component in the two phases is characterized by the distribution coefficient, which is given by the ratio of the concentrations of the component in the fluid phase as represented by two end points of a tie line (Mukhopadhaya, 2000). The increase in solubility due to the addition of co-solvent is the result of additional interactions between the solute and the co-solvents. Considering the interactions possible, these co-solvent effects could be the result of several mechanisms: the addition of a co-solvent generally increases the mixture density, which enhances the solubility as well as physical interactions like dipole–dipole, dipole-induced dipole

and induced dipole interactions (Ting *et al.*, 1993). On the contrary, there are also reports where co-solvents have decreased extraction, both in terms of quality and quantity of total flavanoids and terpenoids. Decreased efficiency can be explained by the phenomenon that, with a co-solvent, higher temperatures are required to reach the increased critical temperature. In addition, more polar substances are extracted together with active compounds as the co-solvent increases their solubility (Yang, Xu and Yao, 2002).

7.3.6 Preparation of plant materials for supercritical fluid extraction and operating conditions

The other crucial parameters in SFE apart from temperature and pressure are the CO_2 flow rate, the particle size of the matrix and the duration of the extraction process. Proper selection of these parameters has the scope of producing complete extraction of the desired compounds in a shorter time. They are connected to the thermodynamics (solubility) and the kinetics of the extraction process in specific raw materials (mass transfer resistances). Proper selection depends on the mechanism that controls the process: the slowest one determines the overall process velocity. The CO_2 flow rate is a relevant parameter if the process is controlled by an external mass transfer resistance or by equilibrium, and the amount of supercritical solvent feed to the extraction vessel; in this case, it determines the extraction rate (Reverchon and Marco, 2006).

Particle size plays an important role in extraction processes controlled by internal mass transfer resistances, since a smaller mean particle size reduces the length of the diffusion of the solvent. However, if the particles are too small, they can give problems of channeling inside the extraction bed. Part of the solvent flows through channels (formed inside the extraction bed) and does not contact the material to be extracted, thus causing a loss of efficiency and yield of the process. As a rule, particles with mean diameters ranging approximately between 0.25 and 2.0 mm are used (Reverchon and Marco, 2006). The optimum dimension can be chosen case by case, considering the moisture content in the matrix and the quantity of extractable liquid compounds that can produce the phenomenon of coalescence among the particles, thus favouring the irregular extraction along the extraction bed. Moreover, the production of very small particles by grinding could produce the loss of volatile compounds. Large particles may require a longer time for extraction, since the process may be controlled by internal diffusion. However, fine powder can speed up the extraction but may also cause difficulty in maintaining a proper flow rate. Chemat *et al.* (2004) used SC-CO_2 to extract *Foeniculum vulgare* volatile oil from fennel fruits with different mean particle sizes, like 0.35, 0.55 and 0.75 mm. They found that a decrease in particle size decreased the total yield of the extracted oil. Therefore, in such cases, some rigid inert materials such as glass beads and sea sand were

placed along with the fine plant powder to maintain a desired permissibility of the particle bed. Preparation of plant materials is another critical factor for SFE of nutraceuticals. Fresh plant materials are frequently used in SFE of nutraceuticals. When fresh plant materials are extracted, the high moisture content can cause mechanical difficulties such as restricting or clogging due to ice formation. Although water is only about 0.3% soluble in SC-CO_2, highly water-soluble solutes would prefer to partition into the aqueous phase, resulting in a low efficiency of SFE. Some chemicals such as Na_2SO_4 and silica gel are mixed with the plant materials to obviate the problem of moisture for SFE of fresh materials (Lang and Wai, 2001). The solubility of a target compound in SC-CO_2 is a major factor in determining its extraction efficiency. The temperature and density of the fluid control the solubility. The choice of a proper density of a SCF such as CO_2 is the crucial point influencing solvency and selectivity, and the main factor determining the extract composition (Cherchi *et al*., 2001). It is often desirable to extract the compound right above the point where the desired compounds become soluble in the fluid so that the extraction of other compounds can be minimized.

In extraction of Chilean hop (*Humulus lupulus*) ecotypes, del Valle *et al.* (2003) found only a marginal increase in the extraction rate when the pressure was above 200 bar and the temperature was 40 °C; rather, an increase in pressure increased the co-extraction of undesirable compounds. Thus by controlling the fluid density and temperature, fractionation of the extracts could also be achieved. For SC-CO_2 extraction of squalene and stigmasterol from the entire plant of *Spirodela polyrhiza*, Choi *et al.* (1997) found that the relative extraction yield of squalene was much higher than that of stigmasterol at 100 bar and 50 or 60 °C. Their results confirmed that SC-CO_2 could selectively extract substances from the plant materials by controlling conditions such as temperature and pressure. The extraction time is the other parameter that determines extract composition. Process duration is interconnected with the CO_2 flow rate and particle size, and has to be properly selected to maximize the yield of the extraction process. A lower molecular weight and less polar compound are more readily extracted during SC-CO_2 extraction since the extraction mechanism is usually controlled by internal diffusion (Cherchi *et al.*, 2001; Poiana, Fresa and Mincione, 1999). Therefore, the extract composition varies with the extraction time.

The importance of nutraceutical products in human health has been well recognized by industries and researchers in the field of supercritical fluids. This fact can be judged by the number of literature emerging in recent years. Advances in supercritical fluid extraction of nutraceuticals and bioactive components have been reflected in several published papers (Martinez, 2008). SFE of bioactives such as carotenoids, polyunsaturated fatty acids (PUFAs), squalene, sterols and tocols (Termelli *et al.*, 2008), tocopherols (Nagesha, Manohar and Udaya Sankar, 2003; Baslingappa *et al.*, 2001), and quinones from black cumin (Suresh Kumar *et al.*, 2010) have been dealt

with. Detailed coverage on extraction of natural tocopherols by SC-CO_2 is presented (Fang *et al*., 2008). Bioactives from fish oils such as omega-3 fatty acids, squalene, diacyl glyceryl ethers and vitamin A (retinol) using SFE have been reviewed (Eltringham and Catchpole, 2008). Bioactives from algae by SC-CO_2 extraction have also been studied (Mendes, 2008). Mangosteen from the pericarp of the mangosteen fruit, supposed to have anticancer and antioxidant properties, has been extracted by SC-CO_2 (Zarena, Manohar and Udaya Sankar, 2012; Zarena, Sachindra and Udaya Sankar, 2012; Zarena and Udaya Sankar, 2009, 2011).

There are important reviews (Shi and King, 2007; Eckert, Knutson and Debenedetti, 1996) dealing with various aspects of supercritical fluid extraction. SFE from food, pharmaceutical, nutraceutical and other natural and biological products has received significant attention in recent years. Rozzi and Singh (2002), Mohamed and Mansoori (2002) and Raventos, Duarte and Alarcon (2002) reviewed applications of supercritical fluids in the food industry. Awasthi and Trivedi (1997), Rizvi (1994) as well as Mukhopadhyay (2000) enumerated extraction techniques for various types of natural materials. King and List (1996) dealt with applications for SFE for lipids and oils. Chen and Ling (2000) as well as Lang and Wai (2001) and Catchpole *et al.* (2004) described applications of SFE technologies for herbal medicine. Apart from these detailed reviews and books, there are several other research publications on supercritical fluids for the extraction of various biological materials (Prieto *et al.*, 2003; Canela *et al.*, 2002; Wong *et al.*, 2001; Senorans *et al.*, 2001; Ibanez *et al.*, 1999; Galan, Nienaber and Schwartz, 1999; Cheng *et al.*, 1999; Ambrosino *et al.*, 1999; Nguyen, Anstee and Evans, 1998; Cheung, Leung and Ang, 1998; Chester, Pinktson and Raynie, 1998; Tsuda *et al.*, 1995; List *et al.*, 1993; Bhaskar, Rizvi and Sherbon, 1993; Peker *et al.*, 1992; Froning *et al.*, 1990). A review on future directions of the process involving SFE has been discussed (Decastro and Carmona, 2000). The list of various bioactive components that have been reportedly extracted with SC-CO_2 and the process parameters, such as temperature, pressure, CO_2 flow, etc., have also been tabulated (Shi and King, 2007). Some studies have focused on other applications, especially with botanical materials and thermally liable substances (Moura *et al*., 2005; Rozzi and Singh, 2002; Walker, Cochran and Hulbert, 1999; Brunner, 1994; McHugh and Krukonis, 1994; Rizvi, 1994; Goto, Sato and Hirose, 1993). Many review articles of supercritical fluid technology exist. Subjects covered include: a general overview (Chester, Pinktson and Raynie, 1998), applications in food industries (Brunner, 2005; Rozzi and Singh, 2002), herbal and natural products (Sovova, 2005; Lang and Wai, 2001), Chinese herbal medicine (Chen and Ling, 2000), pharmaceutical research (Vasukumar and Bansal, 2003; Subramaniam, Rajewski and Snavely, 1997) and biotechnology (Williams and Clifford, 2000).

SC-CO_2 has been used as a solvent to extract natural antioxidants from various plants, including carotenoids, nonpolar tocopherols, terpenoids and polar

phenolic compounds such as flavonoids from oilseeds, plant leaves, labiate herbs and spices (Mukhopadhyay, 2000). Traditional extraction methods such as solvent extraction, aqueous alkaline extraction and steam distillation are not selective, and hence antioxidant extracts often show colour (chlorophyll) and have a strong flavour. Therefore, further purification steps are often required for the extract and final food product to remove unwanted residuals. SC-CO_2 extraction, on the other hand, inherently increases selectivity and allows for fractionation of the extract for high value added substances, such as antioxidants.

Some of the nutraceutical products made by SC-CO_2 extraction and their salient features are:

(a) Extracts of chamomile flowers as anti-inflammatory and antispasmodic bioactive compounds (e.g. sesquiterpene, lactone and matricin) (Scalia, Giuffreda and Pallado, 1999).
(b) Extracts of turmeric for bile preparations – no artefacts such as tolylmethylcarbinol created during steam distillation (Gopalan *et al*., 2000).
(c) Valerian as a sedative preparation – valepotriates obtained are in an undecomposed form having a high yield (>90%) (Zizovic *et al*., 2007).
(d) Wormwood extracts as a carminative, cholagogue and stomachic – removal of toxic β-thujone by fractional extraction from thermally unstable pharmacology active components (Ghasemi *et al*., 2007).
(e) Hydrogenation reactions in supercritical carbon dioxide that are a factor of 1000 times faster than conventional hydrogenation reactions with greater control over trans-isomer formation (Marentis, Hsu and James, 2001).
(f) Saw Palmetto – higher concentrations of phytosterols (active ingredients) (Catchpole *et al*., 2002).
(g) Ginseng – extraction of pesticides without extracting significant quantities of active ingredients (Wang, Chen and Chang, 2001).
(h) Echinacea – more concentrated extract is obtained by SFE than conventional technologies (Catchpole *et al*., 2002).
(i) Feverfew – more concentrated extract is obtained by SFE than conventional technologies (Cretnik, Skergetand Knez, 2005).
(j) Chitin (glucosamine) – able to separate astaxanthin co-product from chitosan using SFE, able to demineralize shells and other processing steps (Lim *et al.*, 2002).
(k) St John's Wort – more concentrated extract is obtained by SFE than conventional technologies (Catchpole *et al*., 2002)
(l) Kava-kava – more concentrated extract is obtained by SFE than conventional technologies (Khorassani, Taylor and Martin, 1999).
(m) Gingko biloba – SFE reduces allergenic compounds in the extract (Yang, Xu and Yao, 2002).

(n) Garlic (allicin) – SFE extract is more concentrated and deodorized in addition to higher yields when compared to conventional technologies (Del Valle, Mena and Budinich, 2008).
(o) Evening primrose oil – more concentrated extract is obtained by SFE than conventional technologies (Favati, King and Muzzanti, 1991).
(p) Rosemary extract – SFE extract is more concentrated and deodorized in addition to higher yields compared to conventional technologies (Ibanez *et al*., 1999).
(q) Grape seed extract – more concentrated extract is obtained by SFE than conventional technologies (Murga *et al*., 2000), and
(r) Separation of essential oil from spices without contamination (Udaya Sankar and Manohar, 1998a).

7.3.7 Commercialization of SFE technology

The early studies on SFE did not just remain as a laboratory curiosity but resulted in the establishment of large commercial enterprises based on SFE using carbon dioxide in Europe and the United States. The first commercial supercritical fluid extraction was performed in Germany in 1978 (Palmer and Ting, 1995) for the decaffeination of green coffee beans. Two years later, Carlton and United Breweries in Australia developed a process for the extraction of hop flavours using liquid carbon dioxide (Palmer and Ting, 1995). Since then, several commercial plants handling several materials have started. Commercial applications of the SFE technology remain limited to a few high-value products due to high capital investment and being a complex operating system. Adoption of the technology is on the rise as a result of advances in processing system, equipment and the realization of producing high-value products with high profitability. Global perspectives for extraction of high-value natural bioactives using SFE technology has been well covered (Udaya Sankar and Manohar, 1998b). A partial list of commercial plants employing SFE technology processing food materials is available. During the last two decades, a large number of industrial plants (about 100) of different capacities were built for batch extractions of solid materials with SCFs. At present, a total number of about 100 extractor vessels larger than 100 litres have been designed for different industrial plants, distributed mainly in Europe, Japan, USA and in other Asian countries (Brunner, 2005). Some of these plants, particularly for hops, tea and coffee, have a production capacity of 15–30 million kg per year.

References

Ambrosino, P., Fresa, R., Fogliano, V., Monti, S.M. and Ritieni, A. (1999) Extraction of azadirachtin-a from neem seed kernels by supercritical fluid and its evaluation by HPLC and LC/MS. *Journal of Agriculture and Food Chemistry*, **47**, 5252–5256.

Awasthi, A. and Trivedi, R.K. (1997) A review on supercritical carbon dioxide extraction of natural products. *Chemical Engingeering World*, **32**(1010), 65–71.

Baslingappa, S.S., Manohar, B., Nagesha, G.K. and Udaya Sankar, K. (2001) Enrichment of tocopherols from deodourized distillate by supercritical fluid carbon dioxide extraction. *Indian Chemical Engineer Section A.*, **43**(4), 283–287.

Bhaskar, A.R., Rizvi, S.S.H. and Sherbon, J.W. (1993) Anhydrous milk fat fractionation with continuous counter current supercritical carbon dioxide. *Journal of Food Science*, **58**(4), 748.

Bicking, M.K.L. (2000) Analytical extractions. In: *Encyclopedia of Separation Science*. Academic Press, pp. 1372.

Brunner, G. (1984) *Bev. Bunsenges Phys. Chem.*, **8**, 887–891.

Brunner, G. (1994) Gas extraction, In: *An Introduction to Fundamentals of Supercritical Fluids and the Application to Separation Processes*. Springer, New York, USA.

Brunner, G. (2005) Supercritical fluids: technology and application to food processing. *Journal of Food Engineering*, **67**, 21–33.

Canela, A.P.R.F., Rosu, P.T.V., Maraques, M.O.M. and Meireles, M.A.A. (2002) Supercritical extraction of fatty acids and carotenoids from microalgae sporulina maxima. *Industrial Engineering and Chemistry Research*, **41**, 3012–3018.

Catchpole, O.J., Perry, N.B., da Silva, B.M.T., Grey, J.B. and Smallfield, B.M. (2002) Supercritical extraction of herbs I: Saw Palmetto, St John's Wort, Kava root, and Echinacea. *Journal of Supercritical Fluids*, **22**(2), 129–138.

Catchpole, O.J., Grey, J.B., Mitchell, K. A. and Lan, J.S. (2004) Supercritical antisolvent fractionation of propolis tincture. *Journal of Supercritical Fluids*, **29**, 97–106.

Chemat, S., Lagha, A., AitAmar, H., Bartels,V., and Chemat, F. (2004) Comparison of conventional and ultrasound-assisted extraction of carvone and limonene from caraway seeds. *Flavour and Fragrance Journal*, **19**, 188–195.

Chen, Y. and Ling, Y. (2000) An overview of supercritical fluid extraction in Chinese herbal medicine: from preparation to analysis. *Journal of Food and Drug Analysis*, **8**(4), 235–247.

Cheng, M.H., Walker, T.H., Hulbert, G.J. and Raman, D.R. (1999) Fungal production of eicosapentaenoic and arachidonic acids from industrial waste streams and crude soybean oil. *Bioresources Technology*, **67**(2), 101–110.

Chester, T.L., Pinktson, J.D. and Raynie, D.E. (1998) Supercritical fluid chromatography and extraction. *Analytical Chemistry*, **70**(12), 301–319.

Cheung, P.C.K., Leung, A.Y.H. and Ang, P.O. Jr. (1998) Comparison of supercritical carbon dioxide and soxhlet extraction of lipids from a brown seaweed. *Journal of Agricultural and Food Chemistry*, **46**, 4228–4232.

Cherchi, G., Deidda, D., Gioannis, B.D., Marongiu, B. and Silvia Porcedda, R.P. (2001) Extraction of *Santolina insularis* essential oil by supercritical carbon dioxide: influence of some process parameters and biological activity. *Flavour and Fragrance Journal*, **16**, 35–43.

Choi, Y.H., Kim, J., Noh, M.J., Choi, F.S. and Yoo K.P. (1997) Comparison of supercritical carbon dioxide extraction with solvent of nonacosan-10-ol, ∝-amyrin acetate, squalene and stigmasterol from medicinal plants. *Phytochemical Analysis*, **8**, 233–237.

Chueh, P.L. and Prasunitz, J.M. (1967*)* Vapour – liquid equilibria at high pressures: vapour phase fugacity coefficients in non-polar and quantum gas mixtures. *Industrial Engineering Chemistry Fundamentals*, **6**, 492–498.

Cocero, M.J. and Garcia, J. (2001) Mathematical model of supercritical extraction applied to oil seed extraction by CO_2+ saturated alcohol: desorption model. *Journal of Supercritical Fluids*, **20**(3), 229–243.

Cretnik, L., Skerget, M. and Knez, Z. (2005) Separation of parthenolide from feverfew: performance of conventional and high-pressure extraction techniques. *Separation and Purification Technology*, **41**(1), 13–20.

Debenedetti, P.G. and Reid, R.C. (1986) Diffusion and mass transfer in supercritical fluids. *AIChE Journal*, **32**, 2034–2046.

Decastro, M.D.L. and Carmona, M.M.J. (2000) Where supercritical fluid extraction is going. *Trends in Analytical Chemistry*, **19**(4), 223–228.

DeFranca, L.F. and Meireles, M.A.A. (2000) Modeling the extraction of carotene and lipids from pressed palm oil (*Elaes guineessis*) fibers using supercritical CO_2. *Journal of Supercritical Fluids*, **18**, 35–47.

del Valle, J.M., Rivera, O., Teuber, O. and Palma, M.T. (2003) Supercritical carbon dioxide extraction of Chilean hop (*Humulus lupulus*) ecotypes. *Journal of the Science and Food Agriculture*, **83**, 1349–1356.

delValle, J.M., Mena, C. and Budinich, M. (2008) Extraction of garlic with supercritical CO_2 and conventional organic solvents. *Brazil Journal of Chemical Engineering*, **25**(3), 535–542.

Eckert, C.A., Knutson, B.L. and Debenedetti, P.G. (1996) Supercritical fluids as solvents for chemical and materials processing. *Nature*, **383**, 313–318.

Eisenbeiss, J. (1964) A basic study of the solubility of solids in gases at high pressure. Report No. 604345, Contract No. Da-18-108-AMC-244(A), Defense Technical Information Center, Cameron Station, Georgia, USA, 6 August.

Eltringham, W. and Catchpole, O. (2008) Processing of fish oils by supercritical fluids. In: Martinez J.L. (ed.), *Supercritical Fluid Extraction of Nutraceuticals and Bioactive Compounds*. CRC Press, New York, USA, p. 141.

Esquivel, M.M., BernardoGil, M.G. and King, M.B. (1999) Mathematical models for supercritical extraction of olive husk oil. *Journal of Supercritical Fluids*, **16**(1), 43–58.

Ewald, A.H., Jepson, W.B. and Rowlison, J.S. (1953) The solubility of solids in gases. *Discussions of the Faraday Society*, **15**, 238–243.

Fang, T., Goto, M., Sasaki, M. and Yang, D. (2008) Extraction and purification of natural tocopherols by supercritical CO_2. In: Martinez, J.L. (ed,), *Supercritical Fluid Extraction of Naturaceuticals and Bioactive Compounds*. CRC Press, New York, USA, p. 52.

Favati, F., King, J.W. and Muzzanti, M. (1991) Supercritical carbon dioxide extraction of evening primrose oil. *Journal of American Oil Chemists' Society*, **68**, 422–427.

Froning, G.W., Wehling, R.L., Coppett, S.L., Pierce, M.M., Niemann, L., Siekman, D.K. (1990) Extraction of chlolestorol and other lipids from dried egg yolk using supercritical carbon dioxide. *Journal of Food Science*, **55**, 95–98.

Galan, M.J., Nienaber, U. and Schwartz, S. (1999) Paprika oleoresin extraction with supercritical carbon dioxide. *Journal of Agricultural and Food Chemistry*, **47**(9), 3558–3564.

Gandhdhara Rao, V.S. and Mukhopadhyay, M. (1988) *Proceedings of the International Symposium on Supercritical Fluids*, 17–19 October, Nice, France, p. 643.

Ghasemi, E., Yamini, Y., Bahramifar, N. and Sefidkon, F. (2007) Comparative analysis of the oil and supercritical CO_2 extract of *Artemisia sieberi*. *Journal of Food Engineering*, **79**(1), 306–311.

Gopalan, B., Goto, M., Kodama, A. and Hirose, T. (2000) Supercritical carbon dioxide extraction of turmeric (*Curcuma longa*). *Journal of Agricultural and Food Chemistry*, **48**(6), 2189–2192.

Goto, M., Sato, M. and Hirose, T. (1993) Extraction of peppermint oil by super-critical carbon dioxide. *Journal of Chemical Engineering Japan*, **26**, 401–407.

Hannay, J.B. and Hogarth, J. (1879) On the solubility of solids in gases. *Proceedings of the Royal Society London*, **29**, 324–326.

Ibanez, E., Oca, A., Murga, G., Sebastian, S. L., Tabera, J. and Reglero, G. (1999) Supercritical fluid extraction and fractionation of different preprocessed rosemary plants. *Journal of Agricultural and Food Chemistry*, **47**, 1400–1404.

Khorassani, M.A., Taylor, L.T. and Martin, M. (1999) Supercritical fluid extraction of Kava lactones from Kava root and their separation via supercritical fluid. *Chromatographia*, **50**(5–6), 287–292.

King, J.W. and List, G.R. (1996) *Supercritical Fluid Technology in Oil and Lipid Technology*. AOCS Press, Champaign, Illinois, USA.

Krishnamurthy, T.P. (2012) Mathematical modeling of unit operations involved on post harvest processing of mango ginger (*Curcuma amado* Roxb). M.Tech. Thesis, Manipal Institute of Technology, Manipal and Central Food Technological Research Institute, Mysore, India.

Lang, Q. and Wai, C. (2001) Supercritical fluid extraction in herbal and natural product studies – a practical review. *Talanta*, **53**, 771–782.

Lim, G.B., Lee, S.Y., Lee, E.K., Haam, S.J. and Kim, W.S. (2002) Separation of astaxanthin from red yeast *Phaffia rhodozyma* by supercritical carbon dioxide extraction. *Biochemical Engineering Journal*, **11**(2, 3), 181–187.

List, G.R., King, J.W., Johnson, J.H., Warner, K. and Mounts, T.L. (1993) Supercritical CO_2 degumming and physical refining of soybean oil. *Journal of the American Oil Chemists' Society*, **70**, 473–476.

McHugh, M.A. and Krukonis, V.J. (1994) *Supercritical Fluid Extraction: Principles and Practice*, 2nd edn. Butterworth-Heinemann, Boston, Massachusetts, USA.

Marentis, R., Hsu, J.T. and James, K. (2001) Supercritical fluid extraction of nutraceutical products. In: 4th Brazilian Meeting on *Supercritical Fluids*, Salvador, Bahia, Brazil, October.

Martinez, J.L. (2008) *Supercritical Fluid Extraction of Nutraceuticals and Bioactive Compounds*. CRC Press, New York, USA.

Martinez, J., Monteiro, A.R., Rosa, P.T.V., Marques, M.O.M. and Meireles, M.A.A. (2003) Multicomponent model to describe extraction of ginger oleoresin with supercritical carbon dioxide. *Industrial Engineering and Chemistry Research*, **42**, 1057–1063.

Mendes, R.L. (2008) Supercritical fluid extraction of active compounds from algae. In: Martinez, J.L. (ed.), *Supercritical Fluid Extraction of Nutraceuticals and Bioactive Compounds*. CRC Press, New York, USA.

Mohamed, R.S. and Mansoori, G.A. (2002) The use of supercritical fluid extraction technology in food processing. *In: Food Technology WMRC*, 1–19 June.

Moura, L.S., Carvalho, R.N., Stefanini, M.B., Ming, L.C. and Meireles, M.A.A. (2005) Supercritical fluid extraction from fennel (*Foeniculum vulgare*) global yield, composition and kinetic data. *Journal of Supercritical Fluids*, **35**, 212–219.

Mukhopadhyay, M. (2000) *Natural Extracts using Supercritical Carbon Dioxide*. CRC Press, New York, USA.

Murga, R., Ruiz, R., Beltrn, S. and Cabezas, L. (2000) Extraction of natural complex phenols and tannins from grape seeds by using supercritical mixtures of carbon dioxide and alcohol. *Journal of Agricultural and Food Chemistry*, **48**, 3408–3412.

Nagesha, G.K., Manohar, B. and Udaya Sankar, K. (2003) Enrichment of tocopherols in modified soy deodorizer distillate using supercritical carbon dioxide extraction. *European Food Research and Technology*, **217**, 427–433.

Nagesha, G.K., Manohar, B., Udaya Sankar, K. (2006) Optimization of enzymatic hydrolysis of triglycerides in soy deodorizer distillate with supercritical carbon dioxide. *Biochemical Engineering Journal*, **29**, 220–226.

Nguyen, U.Y., Anstee, M. and Evans, D.A. (1998) Extraction and fractionation of spices using supercritical carbon dioxide. Paper presented at 5th International Symposium on *Supercritical Fluids*, Nice, France.

Palmer, M.V. and Ting, S.S.T. (1995) Application for supercritical fluid technology in food processing. *Food Chemistry*, **52**, 345–352.

Peker, H., Srinivasan, M.P., Smith, J.M. and Mccoy, B. J. (1992) Caffeine extraction rates from coffee beans with supercritical carbon dioxide. *AIChE Journal*, **38**(5), 761–770.

Perry, R.H. and Green, D.W. (1999) *Perry's Chemical Engineers' Handbook*. McGraw-Hill Companies, Inc., New York, USA.

Pitzer, K.S. and Curl Jr. R.F. (1957) The volumetric and chemothermodynamic properties of fluids. III. Empirical equation for the second virial coefficient. *Journal of the American Chemical Society*, **79**, 2369–2370.

Poiana, M., Fresa, R. and Mincione, B. (1999) Supercritical carbon dioxide extraction of bergamot peels: extraction kinetics of oil and its components. *Flavour and Fragrance Journal*, **14**, 350–366.

Prieto, M.S.G., Caja, M.M., Herraiz, M. and Maria, G.S. (2003) Supercritical fluid extraction of all-trans-lycopene from tomato. *Journal of Agricultural and Food Chemistry*, **51**, 3–7.

Raeissi, S. and Peters, C.J. (2005) Application of double retrograde vaporization as an optimizing factor in supercritical fluid separations. *Journal of Supercritical Fluids*, **33**, 115–120.

Raventos, M., Duarte, S. and Alarcon, R. (2002) Application and possibilities of supercritical CO_2 extraction in food processing industry: an overview. *Food Science and Technology International*, **8**(5), 269–284.

Reid, R.C., Prausnitz, J.M. and Sherwood, T.K. (1977) *The Properties of Gases and Liquids*. McGraw-Hill, London, England.

Reverchon, E. and De Marco, I. (2006) Supercritical fluid extraction and fractionation of natural matter. *Journal of Supercritical Fluids*, **38**, 146–166.

Rizvi, S.S.H. (1994) *Supercritical Fluid Processing of Food and Biomaterials*. Blackie Academic and Professional, Glasgow, Scotland.

Rowlison, J.S. (1969) *Liquid and Liquid Mixtures*, 2nd edn. Butterworth, London, England.

Rowlison, J.S. and Richardson, J.M. (1959) In: Priogine, I. (ed.), *Advances in Physics*, vol. **II**. Interscience, New York, USA, p. 85.

Rozzi, N. L. and Singh, R.K. (2002) Supercritical fluids and food industry. *Comprehensive Review, Food Science and Food Safety*, **1**, 33–44.

Scalia, S., Giuffreda, L. and Pallado, P. (1999) Analytical and preparative supercritical fluid extraction of Chamomile flowers and its comparison with conventional methods. *Journal of Pharmacology and Biomedical Analysis*, **21**(3), 549–558.

Schneider, G.M. (1978) *Chemical Engineering Thermodynamics*, vol. **2**, Chapter 4. Specialist Periodical Reports, Chemical Society, London, England.

Senorans, F.J., Rodriguez, A.R., Cavero, S., Cifuentes, A., Ibanez, E. and Aeglero, G. (2001) Isolation of antioxidant compounds from orange juice by using counter current supercritical fluid extraction. *Journal of Agricultural and Food Chemistry*, **49**, 6039–6044.

Shi, J. and King, J.W. (2007) *Functional Food Ingredients and Nutraceuticals: Processing Technologies*. CRC Press, Taylor & Francis Group, Florida, USA.

Sovova, H. (1994) Rate of the vegetable oil extraction with supercritical CO_2 modeling of extraction curves. *Chemical Engineering Science*, **49**(3), 409–414.

Sovova, H. (2005) Mathematical model for supercritical fluid extraction of natural products and extraction curve evaluation. *Journal of Supercritical Fluids*, **33**(1), 35–52.

Street, W.B. (1983) Phase equilibria in fluid and solid mixtures at high pressure. In: Paulaitis, M.E., Penninger, J.M.L., Gray, R.D., David, P. (eds), *Chemical Engineering at Supercritical Fluid Conditions*. Ann Arbor Science, Michigan, USA, p. 3.

Subramaniam, B., Rajewski, R.A. and Snavely, K. (1997) Pharmaceutical processing with supercritical carbon dioxide. *Journal of Pharmaceutical Science*, **86**(8), 885–890.

Suresh Kumar, T.V, Pattekhan, H.H., Divakar, S. and Udaya Sankar, K. (2010) Chemical composition of essential oil from *Nigella sativa* L. seeds extracted by supercritical carbon dioxide. *Journal of Food Science and Technology*, **47**(6), 598–605.

Tan, C. and Liou, D. (1989) Modeling of desorption at supercritical conditions. *AIChE Journal*, **35**(6), 1029–1031.

Termelli, F., Saldaña, M.D.A, Moquin, P.H.L. and Sun, M. (2008) Supercritical fluid extraction of specialty oils. In: Martinez J.L. (ed.), *Supercritical Fluid Extraction of Nutraceuticals and Bioactive Compounds*. CRC Press, New York, USA, p. 52.

Ting, S.S.T., Macnaughton, S.J., Tomasko, D.I. and Foster, N.R. (1993) Solubility of Naproxen in supercritical carbon dioxide with and without co-solvents. *Industrial Engineering and Chemistry Research*, **32**, 1471–1481.

Treybal, R.E. (1986) *Mass Transfer Operations*, 3rd edn. McGraw-Hill, NewYork, USA.

Tsekhanskaya, Y.V., Iomter, M.V. and Muskina, E.V. (1964) Solubility of diphenylamine and naphthalene in carbon dioxide under pressures. *Russian Journal of Physics and Chemistry*, **36**, 1177.

Tsuda, T., Mizuno, K., Ohshima, K., Kawakishi, S. and Osawa, T. (1995) Supercritical carbon dioxide extraction of antioxidant components from tamarind seed coat. *Journal of Agricultural and Food Chemistry*, **43**, 2803–2806.

Udaya Sankar, K., Manohar, B. and Chokkalingam (1986) A note on supercritical carbon dioxide decaffeinated coffee. *Journal of Food Science and Technology*, **23**, 326–328.

Udaya Sankar, K. and Manohar, B. (1994) Mass transfer phenomena in supercritical carbon dioxide extraction for production of spice essential oils. In: Rizvi, S.S.H. (ed.), *Supercritical Fluid Processing of Food and Biomaterials*. Chapman and Hall, Bishopbriggs, Glasgow, Scotland, pp. 44–53.

Udaya Sankar, K. and Manohar, B. (1998) Extraction of essential oils using supercritical carbon dioxide. In: *Proceedings of the Symposium on the Supercritical Fluids*, Perrut, M. (ed.), vol. **2**, Vandeouvre, France, pp. 807–814.

Valcárcel, M. and Tena, M.T. (1997) Applications of supercritical fluid extraction in food analysis. *Fresenius Journal of Analytical Chemistry*, **358**, 561–573.

Vasukumar, K. and Bansal, A.K. (2003) Supercritical fluid technology in pharmaceutical research. *CRIPS*, **4**(2), 8–12.

Walker, T.H., Cochran, H.D. and Hulbert, G.J. (1999) Supercritical carbon dioxide extraction of lipids from *Pythium irregulare*. *Journal of the American Oil Chemists' Society*, **76**(5), 595–602.

Wang, H.C., Chen, C.R., Chang, C.J. (2001) Carbon dioxide extraction of ginseng root hair oil and ginsenosides. *Food Chemistry*, **72**(4), 505–509.

Wang, L., Weller, C.L. (2006) Recent advances in extraction of nutraceuticals from plants. *Trends in Food Science and Technology*, **17**(6), 300–312.

Williams, D.F. (1981) Extraction with supercritical gases. *Chemical Engineering Science*, **36**(11), 1769–1788.

Williams, J.R. and Clifford, A.A. (2000) *Supercritical Fluid Methods and Protocols*. Humana Press, Totowa, New Jersey, USA.

Wong, V., Wyllie, S.G., Cornwell, C.P. and Tronson, D. (2001) Supercritical fluid extraction of monoterpenes from the leaves of tea tree. *Molecules*, **6**, 92–103.

Yang, C., Xu, Y.R. and Yao, W.X. (2002) Extraction of pharmaceutical components from ginkgo biloba leaves using supercritical carbon dioxide. *Journal of Agricultural and Food Chemistry*, **50**(4), 846–849.

Young, C.L. (1978) *Chemical Engineering Thermodynamics*, vol. **2**, Chapter 3. Specialist Periodical Reports, Chemistry Society, London, England.

Zarena, A.S., Manohar, B. and Udaya Sankar, K. (2012) Optimization of supercritical carbon dioxide extraction of xanthones from mangosteen pericarp by response surface methodology. *Food and Bioprocess Technology*, **5**(4), 1181–1188.

Zarena, A.S., Sachindra, N.M. and Udaya Sankar, K. (2012) Optimisation of ethanol modified supercritical carbon dioxide on the extract yield and antioxidant activity from *Garcinia mangostana* L. *Food Chemistry*, **130**(1), 203–208.

Zarena, A.S. and Udaya Sankar, K. (2009) Supercritical carbon dioxide extraction of xanthones with antioxidant activity from *Garcinia mangostana*: characterization by HPLC/LC–ESI-MS. *Journal of Supercritical Fluids*, **49**(3), 330–337.

Zarena, A.S. and Udaya Sankar, K. (2011) Xanthones enriched extracts from mangosteen pericarp obtained by supercritical carbon dioxide process. *Separation and Purification Technology*, **80**(1), 172–178.

Zizovic, I., Stamenić, M., Ivanović, J., Orlović, A., Ristić, M., Djordjević, S., Petrović, S.D. and Skala, D. (2007) Supercritical carbon dioxide extraction of sesquiterpenes from valerian root. *Journal of Supercritical Fluids*, **43**(2), 249–258.

Zosel, K. (1978) Separation with supercritical gases. *Angewandt Chemical International Edition England*, **17**, 702–710.

8
Baking

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8.1 Introduction

Bakery products are known to people from ancient times and home baking was the common practice. The concept of baking food or baking as an industry is as old as human civilization. Consumer demand for ready-to-eat processed foods with a better shelf-life, taste, nutritional quality and health benefits has increased thoughout the world. Bakery products are the important items that can satisfy most of these requirements. Bakery products include varieties of bread, such as yeast leavened breads (like white bread) and unleavened breads (such as tortilla, *chapati* and *tandoori roti* and sweet products such as cookies, biscuits, cakes and muffins).

Baking is a thermal process that is carried out under high temperatures. Baking is the key unit operation in a bakery, wherein an intermediate dough or batter develops the desired product characteristics including volume, colour, texture and flavour. Several physical, chemical, biochemical and rheological changes that take place during the baking process include volume expansion, vaporization of water, formation of porous structure, denaturation of protein, gelatinization of starch, colour formation due to the browning reaction and crust formation. An understanding of these changes is highly useful in the development of a complete mathematical model of the process of baking. Baking is thus a complex process wherein both heat and mass transfer take place simultaneously and convert the dough or batter into rigid products. High quality of a bakery product is possible to obtain when the quantity of good quality ingredients are optimized along with the baking process. In the baking process, heat is mainly transferred by convection from the heating

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medium and by radiation from the oven walls to the product. This is followed by conduction heat transfer from the peripheral to the geometric centre of the product. The product properties, such as specific heat, thermal conductivity, thermal diffusivity and moisture diffusivity, are important to predict the temperature and moisture distribution in the product during the baking process (Sablani *et al.*, 1998; Rask, 1989). The application of the baking process to manufacture several bakery products, the role of ingredients and process, and the changes that take place during the baking of bread, biscuits and cakes will be discussed in this chapter.

8.2 Bread

Baked products have a long history of production since humans first learnt how to bake cereal grains to improve their palatability and digestibility. It is possible that the first baked product was developed in the ancient Middle East. The technology of baking spread rapidly wherever wheat and other cereal grains could be grown. Bread is a 'balanced' product, providing a good source of energy, protein, vitamins (particularly the B group vitamins), minerals and dietary fibre. The major ingredients of bread include flour, water, yeast and salt. Optional ingredients that may be added include malt flour, soya flour, dough conditioner, enzyme, sugar, yeast, milk and milk products, fat and fruit. Varieties of bread differ in size, shape, colour, texture and flavour. For example, the soft roll is a type of bread characterized by its softness, sweetness and fine texture (Hui, 2006).

8.2.1 Classification of bread

Breads are broadly classified into leavened and unleavened types. In leavened breads, leavening agents like bakers yeast (*Saccharomyces cerevisiae*) or baking soda (sodium bicarbonate), acids and inert fillers are used to expand bread by incorporating carbon dioxide. Examples of leavened bread include the most common white bread, different styles of brown bread, whole grain (whole meal) bread and many types of gourmet bread (mixed grain and fruits). Unleavened bread is often flat and dense due to the absence of gas-producing agents, which include pancake, muffins and Arabic bread.

Ingredients and their role

Flour Wheat flour is unique among the cereal flours in that when mixed with water in appropriate proportions it forms an elastic dough. This phenomenon of gluten formation is used in the preparation of baked products such as breads (yeast breads), quick breads, cakes and pastries; a strong network is formed that traps the gas produced by yeast or baking powder to offer a spongy structure. The protein content of wheat flour is thus one of the most important

parameters affecting bread quality. An increase in protein content leads to an increase in the gas-retention properties of the dough and therefore increases bread volume. The main wheat proteins of interest in baking are classically divided into two fractions, referred to as gliadins and glutenins, and both contribute to flour quality and dough rheological properties. Glutenin controls the elasticity of the gluten and the time required for dough development, while gliadin is responsible for the loaf volume (Shakuntala and Shadaksharaswamy, 2008). Properly developed gluten makes the dough easy to handle, either by hand or machine, and permits a good expansion during fermentation and at the early stage of baking.

Bread flours are milled from blends of hard winter and spring wheat, and their moisture, protein and ash contents, quality of starch and particle size are controlled. The flour quality requirements of bread, biscuits and cake are given in Table 8.1.

Water An important function of water is gluten formation. The amount of water absorbed to make dough of standard consistency depends upon the proportion of protein and damaged starch. Water brings about the partial gelatinization of starch and the ingredients into intimate contact so that the complex reaction of bread making can take place. Water also dissolves sugars and salt, and serves as a dispersion medium for yeast cells. In general, water of a medium hardness (50 to 100 ppm) with a neutral or slightly acidic pH is preferred for dough making. Water that is too soft can result in sticky doughs

Table 8.1 Flour quality requirements for bread, biscuit and cake

Quality parameter	Bread	Biscuit/cookie	Cake
Protein (%)	12–13	7–8: Wire-cut cookies 8–9: Rotary moulded 8–10: Crackers/ semi-sweet	7–8: Normal 9–9.5: Fruit cake
Gluten (%)	~10	~8	7–8
Ash (%)	0.42–0.48	0.45	0.4
Colour grade	3–4	3–4	~3
Damaged starch (%)	7–9	Low	Minimum
Falling number (s)	350	400–450	450–550
Water absorption (%)	60–65	54–56	54
Sedimentation value (mL)	30–40	20	<20
Farinograph stability (min)	6–8	2–3	2–3
Extensograph 'R' value (BU)	550–700	200–300	200
Extensograph 'E' value (mm)	150–200	>200	200
Granulation	Medium	98–100% (220 μm) 70–80% (180 μm) 20–30% (130 μm)	Finer

because of the absence of gluten tightening minerals. Too hard water may retard fermentation to a certain extent by toughening the gluten. Alkaline water affects the functioning of yeast and flour enzyme (Matz, 1960). Gelatinization during baking plays a significant role in the formation of the product structure and the changes that subsequently occur as the product is cooled and stored. The impact of damage to the starch granules extends beyond increasing the water absorption capacity of the flour. High levels of starch damage in white flours can lead to the loss of bread volume. Cauvain and Young (2006) discussed the effect of excessive damaged starch in the context of the Chorleywood bread process (CBP), where it leads to a more open cell structure and greying of bread crumb. This relationship between starch damage and alpha-amylase activity has profound implications for baked product quality, especially for bread and fermented goods. The release of water from starch leads to softening of the dough, which may cause processing problems. The presence of high levels of dextrins can offer problems during slicing of bread (Cauvain and Young, 2006).

Yeast Bakers yeast (*Saccharomyces cerevisiae*) is used to produce carbon dioxide in the manufacture of bread. It acts on simple sugars to produce both carbon dioxide and alcohol (Williams and Pullen, 1998). Yeast used for leavening is marketed in two forms: compressed yeast and active dry yeast. Compressed yeast has a moisture content of 72% and is the most active form of yeast for bread making, but is highly unstable. The shelf-life of compressed yeast is only about five weeks and that of dry yeast is two years.

Carbon dioxide has an important contribution for the expansion of baked products and imparts significantly to changes in texture and eating quality. The alcohol is driven off during baking. Gradual production of carbon dioxide is preferable, because the film-forming property of gluten is also developed gradually as the dough is being hydrated and mechanically kneaded. The production of carbon dioxide and alcohol from sugars is a temperature-sensitive reaction, the optimum temperature being 40–43 °C. The heat of the baking operation eliminates the yeast and inactivates enzymes; thus, fermentation and release of carbon dioxide ceases (Potter and Hotchkiss, 1995). Bakers yeast acts on simple sugars like sucrose by hydrolysing it into glucose and fructose by means of the enzyme invertase. The yeast then immediately ferments glucose and later it proceeds to ferment fructose.

Salt (sodium chloride) Salt is used for a variety of purposes in the manufacture of baked products; it contributes to flavour development. The ionic nature of salt controls water activity and therefore retards the growth of molds (Cauvain and Young, 2006). Salt limits the activity of yeast in dough. The lower the level of salt in the dough, the lower the yeast level will be to maintain a given proof time (Williams and Pullen, 1998). The presence of salt also affects gluten formation during the dough-making stage.

Sugars Added sugar is not essential for bread making. A small quantity of sugar is present in the flour, which is readily used up by the yeast. However, if no sugar is added, fermentation is slow because sugar is needed for yeast activity, which must be formed by the action of amylases on starch. Excess sugar will retard the fermentation by the osmotic effect of dissolved solutes on yeast cells, and it also interferes with gluten formation. The brown colour of the crust of bread is due to the Maillard reaction between the amino acid of protein and reducing sugar during baking. Sugar also possesses moisture-retaining properties in baked goods. Increasing sugar level lowers the water activity of a product and has a significant effect on the shelf-life.

Shortenings Oils and fats have been added to modify the mouth-feel of baked products. Shortenings in bread have a tenderizing effect. The main role of solid fat in bread is stabilization of gas bubbles formed due to yeast fermentation in dough, which improves the gas-retention properties and is usually manifested as improved oven spring (height between the dough entering the oven and the baked bread). It inhibits gas bubble coalescence and offers a finer structure and softness of crumb at higher levels of addition.

Emulsifiers Emulsifiers belong to the general class of surface-active agents and are able to form an emulsion with two liquids that are normally immiscible. Emulsifiers are widely used as ingredients in shortening for bread. Monoglycerides and diglycerides of fatty acids are the most widely used emulsifiers, and these are able to form complexes with the starch, which slow down the retrogradation process in the baked products during storage (Pateras, 1998). Emulsifiers improve dough handling properties, the rate of hydration and water absorption, increase greater tolerance to resting time, shock and fermentation, and improve crumb structure (finer and closer grain, brighter crumb, increased uniformity in cell size). They also emulsify fats, improve gas retention, have better oven spring, increased loaf volume and bread shelf-life (Stampfl and Nersten, 1995). A wide range of emulsifiers is used in the baking industry. The major differentiation is between crumb softeners, which improve the softness of baked products, and act as dough conditioners, which improve dough properties. The most commonly used emulsifiers are glycerol monostearate and lecithin. One of the most common emulsifiers for water-in-oil emulsions is monoglycerides of fatty acid. Glyceryl monostearate is used as a bread improver; it has a dominating effect on softness but less effect on loaf volume, resulting in a fine crumb with considerable elasticity. The action of glyceryl monostearate is based on retarding starch retrogradation.

Dough conditioners A scientific explanation for the loaf volume increase and improved crumb structure due to emulsifiers cannot be offered, but a theory based on the emulsification or lubricating effect appears promising. Flour,

water, salt and yeast are mixed into a complex dough, consisting of starch that is held together by gluten strands and water. These gluten strands form a network. The addition of fat decreases the friction between the gluten strands themselves and between the gluten strands and starch particles, which results in improved viscoelastic properties of the dough. Both these effects contribute to improved gas retention. An emulsifier provides good emulsification of fat in dough and dispersion of the fat as a film between gluten strands and starch particles. The unstable starch–water dispersion becomes stabilized as the hydrated starch granules repel each other. All these effects give a good 'conditioned' dough or 'strengthened' dough, which results in increased loaf volume, a fine and regular crumb structure and crumb grain, and softer bread (Hui, 2006).

Enzymes Small amounts of other ingredients are added to modify the properties of commercial bread. These include enzymes, hydrocolloids, dough improvers, etc. Wheat flour contains a wide range of enzymes, mostly located in the aleurone layer and germ. The addition of enzymes is commonly used to modify the dough rheology, gas retention and crumb softness in bread manufacture (Williams and Pullen, 1998), and for the reduction of acrylamide formation in bakery products (de Boer, Heermans and Meima, 2005).

Amylases, proteases and oxidoreductases are the common enzymes used in baked products. Alpha amylases are used to improve the gas-retention properties of fermented dough, which leads to improvement in product volume, softness and crust colour, increases fermentable sugars and enhances flavour. Proteolytic enzymes act on the proteins of wheat flour and reduce gluten elasticity, thereby causing the dough to become softer and more extensible.

Hydrocolloids In the baking industry, hydrocolloids are gaining increasing importance for improving the bread-making process. They improve dough handling properties, quality of fresh bread and extend the shelf-life of stored bread (Rosell, Rojas and Benedito de Barber, 2001a). Some of the examples of hydrocolloids used are carboxymethylcellulose, xanthan, sodium alginate and hydroxypropylmethylcellulose.

Bread-making process Several developments in bread making have taken place in recent years. These developments are aimed at (a) reducing processing time, (b) improving bread quality and (c) making the production continuous. Bread preparation includes mixing, fermentation, dough make up, proofing, baking, cooling, slicing and wrapping. The major processing methods in bread production are as follows.

Straight dough method All the ingredients are mixed at one time and allowed to ferment. After resting the dough for about two hours, it is knocked back to

remove the gas evolved. It is then allowed to rise again for one hour, which is called first proofing. The dough is then moulded and allowed to rest in the baking pan for 45 to 60 min for the final proofing and is then baked (Shakuntala and Shadaksharaswamy, 2008). Among all the bread-making methods, the straight dough method is simplest and most commonly practised. Based on differences in fermentation time, the straight dough method can further be divided into two types: straight dough and no-time dough. Breads made through the straight dough method need a shorter fermentation time and have less fermentation loss. Breads made by the speedy method (no-time dough) are dense in texture and rough due to insufficient fermentation and have a significantly decreased shelf-life; the overall quality of the bread is also not ideal.

Sponge and dough method The sponge process involves two-stage mixing. A portion of flour and water and yeast are mixed to create the sponge dough. Occasionally, depending on the fermentation time and the flour strength, small amounts of sugar, yeast and improvers are added to strengthen the sponge dough. After pre-fermentation, the dough is mixed for a second time with other ingredients such as water, milk powder, salt, sugar and fats and oils to create the main dough. After the main dough completes its extended fermentation, it proceeds to dividing, rounding and other steps in the bread-making process. Sufficient fermentation of the dough increases the bread volume; the texture of bread is finer and soft, and it possesses a unique flavour and aroma; the staling rate also slows down.

Based on the different proportions of flour added, sponge processes are divided into two broad categories: plastic sponge (or solid sponge) and liquid sponge. In the plastic sponge process, the dough is made using over 55% of the total flour in the formula and the proportion of water needed must be 58–62% of the plastic flour. In the liquid sponge process, the amount of flour usually used in making the liquid sponge dough is about 30–50% of the total flour in the formula. In cases where the amount of flour used is smaller than 30% of the total flour, 1% of sugar can be added to help fermentation. The amount of yeast used is around 0.5% of the amount of the liquid sponge flour and the amount of water to be added should be at least 1:1 (water:flour) in proportion with the liquid sponge flour, but can be as high as 1.25:1. The general proportion is 1.05 to 1.15:1. Since the proportion of water added to the liquid sponge dough is much higher than that added to the plastic sponge dough, the mixture does not form dough. Instead, it exists in a thick colloidal liquid form and is called 'liquid sponge' (Hui, 2006).

The Chorleywood method is designed for wheat flours that are strong in nature and meant for production of high loaf volume bread. The yield of bread in this method is about 4% higher than the conventional method. The process is mostly a batch process and requires considerable high-speed mechanical mixing in order to develop the required dough structure within a short time. Bread produced by this method produces a bland flavour.

8.2.2 Principles of baking

The different unit operations for processing of bread, biscuit and cakes are shown in Figure 8.1 and are discussed in subsequent sections.

Mixing Most of the characteristics of the final products are determined directly or indirectly during the mixing stage. If the dough is undermixed or overmixed, the handling properties of the dough will be different. The metabolism of yeast may be changed if overmixed. Consequently, formation of yeast flavour substances in the crumb may be prohibited (Zehentbauer and Grosch, 1998). Mixing is normally designed to achieve a target energy input into dough or a target final dough temperature. The importance of

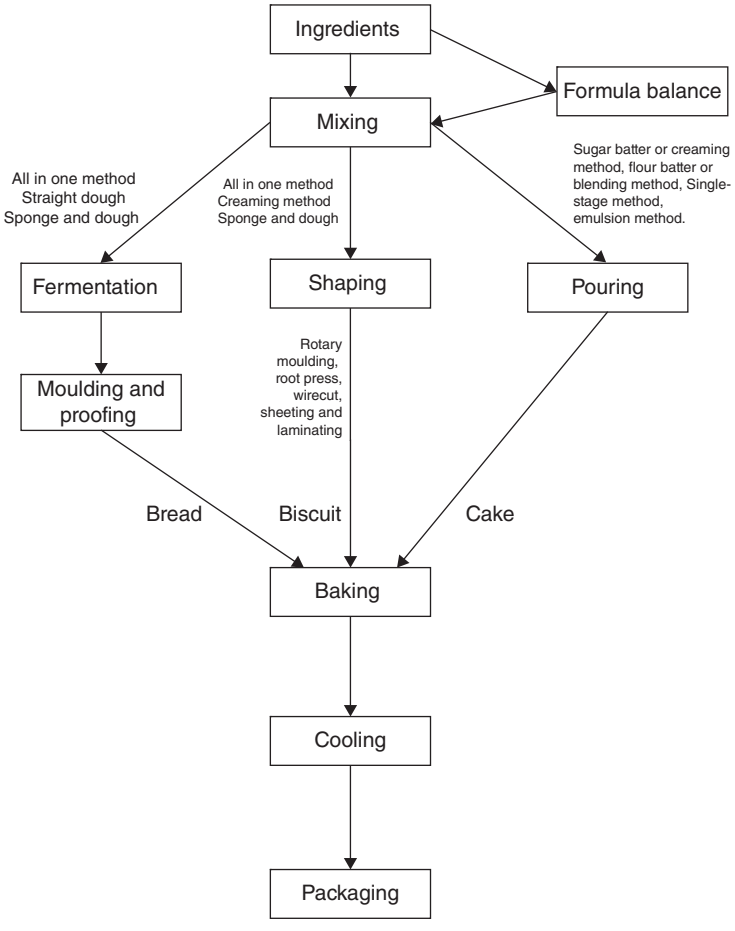

Figure 8.1 Different unit operations for processing of bread, biscuit and cake

the dough temperature also cannot be underestimated. If the temperature is inappropriate, the fermentation rate will be faster or slower, and that in turn will influence the final volume of the bread and colour of the crust. Mixing time influences texture and grain of the crumb. Mixing is normally a discontinuous step in the sponge dough method but is a continuous process in the straight dough method.

Objective of mixing phase Multiple objectives are achieved during mixing; these are the uniform incorporation of all ingredients, hydration of the flour and the other dry ingredients and development of gluten (Hui, 2006).

Fermentation Fermentation in bread making is the process by which the well-mixed ingredients for bread making are converted under controlled temperature and humidity for an appropriate time, to soften and expand dough, with changes in both structural and rheological properties. The yeast converts the available carbohydrates (sugars) to carbon dioxide that enables dough volume expansion and bread softness. The ingredients in the dough undergo both physical and chemical changes during the fermentation process. The physical changes include an increase in volume and temperature of the dough and changes in consistency. Chemical changes include the lowering of pH from 5.5 to 4.7 due to the formation of acids like acetic acid, thus providing the bread with flavour and aroma. In optimally fermented dough, both gas production capacity and gas retention capacity should coincide. When both peaks are reached at the same time, bread will have the largest loaf volume and good texture.

Fermentation time Fermentation time for bread is mainly related to the amount of yeast used. The more yeast used, the less is the fermentation time needed. Besides the amount of yeast used, the mixing level of the dough, dough temperature, the temperature and humidity of the fermenting environment and the amount of improvers and enzyme used may influence the fermentation condition of the bread. Therefore, the fermentation time mainly depends on the amount of yeast used.

Factors influencing fermentation Many factors influence dough fermentation, including the components of flour, salt, sugar, yeast, water, improvers, the degree of dough mixing, dough temperature, fermentation time, and environmental temperature and humidity control.

Dough make-up The fermented bulk dough is divided into individual dough pieces of proper weight, which, when moulded, proofed and baked, will yield the desired baked product. Dough make-up includes dividing, rounding, intermediary proofing and moulding. The dough is divided into loaf-sized pieces

with the help of a divider. Dough is usually divided on the volumetric basis; dividing should be done within the shortest time in order to ensure a uniform weight. Rounding is the process in which the pieces of dough are shaped into round balls with smooth unbroken skin over its entire surface. If the divided dough piece is irregular in shape, the gas can readily diffuse and the gluten structure becomes disoriented and becomes unsuitable for moulding. Rounding is done in order to ensure the external surface smoothness and, for the dry interior of dough pieces with a relatively thick and continuous skin around them, to reorient the gluten structure.

Intermediate proofing is the process of giving the rounded dough pieces a short rest period (about 12–15 min) to recover from the effects of the dividing and rounding machines. In intermediate proofing, the dough becomes larger in volume due to gas accumulation and the skin is firmer and drier, making the dough pieces more pliable and extensible. Upon completion of the intermediate proofing period, the dough pieces are moulded into the desired shape. One main principal objective of the moulding process is to expel carbon dioxide as much as possible. In the moulder, the dough passes through three distinct stages. Flattening is done in the head rollers of the moulder. The sheeting rollers sheet the dough into a fit piece of dough and the curling rollers and thread rollers twirl each piece of sheeted dough to give it a cylindrical shape. In the next process, the drum or pressure plate rolls and seals the loaf into its final form.

Panning Pan proofing is the process of rolling the panned and racked moulded dough pieces quickly into the dough proofing cabinet, which is well insulated and maintained at a temperature of 35–37 °C and a relative humidity of 85%. The panning process is to be so adjusted that it will deposit the dough loaf into the pan with the seam down. This will prevent the subsequent opening of the seam during final proofing and baking, thereby minimizing the appearance of rough and irregular top crust surfaces.

Proofing The purpose of proofing is to relax the dough from the stress received during the moulding operation. Proofing is done to bring about the recovery of those dough properties that have been lost during dough make-up and to facilitate the production of gas in order to give a good volume to the bread and to change the tough bucky gluten to a good mellow extensible character. Temperature, humidity and time play major roles during proofing. The proofing temperature selected is influenced by factors such as flour strength, dough formulation, dough conditioners and shortenings used, and the degree of fermentation. Relative humidity may vary from 75 to 90%. Lower humidity tends to promote the formation of an excessively dry skin on the proofing dough, which will restrict its optimum expansion and desired crust colouration during baking. Excessive humidity causes moisture condensation on the dough and results in a tough crust and blisters in the

finished bread. Dough entering the oven usually has a lower temperature than that of the equipment. Typically, dough temperatures will vary within the range of 25 and 32 °C, depending on the bread-making method employed and the temperature of the environment. In warm seasons, the dough temperature may rise to as high as 35–36 °C. Whatever may be the actual temperature of the dough pieces, it is usually lower than that of the oven at the point of entry. The dough temperature in the oven rises quickly at the surface and more slowly at the centre. The shape and form of the dough piece has a direct impact on the rate at which the dough temperature will rise. Dough pieces with thin cross-sections tend to proof more rapidly and the yeast level may be lowered in order to extend the proof time. The changes in dough rheology that occur with time are helpful in controlling expansion in the oven. The proof time is between 55 and 65 min. The actual proof time will vary depending on the dough character. Poor proofing results in loaves with a pale crust colour, coarse grain, poor texture, impaired keeping quality and a flavour with an acid overtone. Underproofing usually yields small loaf volumes, shell tops, a foxy red crust colour and occasional bursting at the sides.

Baking Baking is generally defined as the process in which products are baked through a series of zones, with exposure to different time periods, temperatures and humidity conditions (Hui, 2006). The final step in bread making is the baking process in which the dough piece is transformed into a light, readily digestible and flavourful product under the influence of heat. Within this baking process, the natural structures of the major dough constituents are altered irreversibly by a series of physical, chemical and biochemical interactions. Several apparent phenomena are caused by oven heat; these involve the expansion in volume, the formation of an enveloping crust, the inactivation of yeast and enzymatic activities, the coagulation of the flour protein and partial gelatinization of flour starch. Meanwhile, the formation of new flavour substances, such as caramelized sugars, pyrodextrins and a broad range of aromatic compounds also accompanies this process.

8.2.3 Changes due to baking

Physical changes The first observable change at the beginning of the baking stage is the formation of a thin and initially expandable surface skin; its elasticity is a direct function of the moisture content of the oven atmosphere. The rise in temperature of baking products speeds up the enzymatic activities and yeast growth at this stage. These two phenomena rapidly increase carbon dioxide production and keep the loaf expanding; this expansion is commonly called ‘oven rise’ or ‘oven spring’. The release of dissolved gases takes place when the internal temperature is around 50 °C. In the following approximately 13 min, the second and third stages of baking occur. The temperature of the

crumb rises at a rate of 5.4 °C per min during the second stage. When the temperature of the crumb reaches about 98.4–98.8 °C, the third stage of baking starts immediately, where the liquids having low boiling points are converted into vapours. The alcohol formed by yeast action further increases the internal pressure, which helps in further oven rise. The temperature of the crumb remains fairly constant at this stage.

Chemical and biochemical changes The reaction rates of starch gelatinization and protein coagulation reach their maximum. The temperature of the crust reaches 150–205 °C and brown colouration begins to appear. In the final stage, the oven temperature remains between 221 and 238 °C. The cell walls of the loaf start firming up and crust colour develops to a desirable extent. In this final stage, certain organic substances volatilize; this is commonly referred as 'bake-out' loss (Prouty, 1965). Yeast generates more carbon dioxide and alcohol in the dough until the thermal death point is reached at 60 °C, which contributes to further expansion of the dough.

Starch gelatinization During the baking process, the starch granules begin to swell at a temperature of about 40 °C. When the temperature reaches a range of about 50–65 °C, the viscoelastic properties of the dough are replaced by fluidity. In the initial stage of gelatinization, the starch granules swell by absorbing both free water and water held by the proteins of the dough. However, a large portion of granules remains intact until the end of gelatinization due to limitation of the water supply (Sansstedt, 1961). The extent of starch gelatinization is influenced by water availability, temperature and the duration of its action on starch. In general, there is a higher degree of starch gelatinization between the crumb layers and the crust than in the centre of the product due to longer exposure at high temperatures (Yasunaga, Bushuk and Irvine, 1968).

Protein denaturation The gluten-forming proteins bind approximately 31% of the total water absorbed by the dough. They contribute to the formation of the dough structure by providing the matrix in which small starch granules are embedded (Pomeranz, Meyer and Seibel, 1984). The proteins begin to undergo thermal denaturation when the temperature of the crumb reaches about 60–70 °C. The denatured proteins start losing their water-binding ability and release water from protein to starch to cause starch gelatinization. On the other hand, when the temperature of the dough rises above 74 °C, the gluten films surrounding the individual gas vacuoles are denatured by heat and are transformed into a semi-rigid structure by interaction with the swollen starch.

Enzyme activity For every 10 °C rise in temperature, the amylases accelerate their hydrolysis of starch. As the temperature rises, thermal inactivation

of enzymes also commences. In the early stage of baking, the amylases contribute two effects in the baking process; these are (1) the amylases attack the starch structure and cause the dough to become more fluid and, hence, promote dough expansion and (2) the starch is broken into small molecules and increases the levels of both dextrins and maltose to be fermented by yeast. Inadequate amylase reactions may induce product defects. The product volume is reduced with low levels of amylase activity; conversely, excessive amylase activity produces over-expansion of the loaf and may cause the loaf to collapse (Marston and Wannan, 1976).

Sugar caramelization When sugar is heated beyond 170 °C, it polymerizes to form coloured substances called caramels. Caramelization takes place in the crust because of the increase in the crumb temperature; it imparts a distinct flavour to the baked product.

Maillard reaction The Maillard reaction is a thermal chemical reaction, which occurs between proteins and carbohydrates during baking to produce melanoidins. It imparts a typical colour and flavour. Lane and Nursten (1983) studied over 400 model systems involving mixtures of twenty-one amino acids and eight sugars that had been heated under different conditions of temperature and humidity, and identified the odours produced from these systems. They have indicated that the odour of bread crust, biscuit, cake and so forth are produced by heating the carbohydrates with the amino acids such as arginine, glutamine, histidine, lysine, proline, serine, threonine and tyrosine at temperatures of 100–140 °C for 0.5–4.0 min. The three stages of Maillard reaction are shown in Figure 8.2.

Proper cooling of bread is required prior to slicing and packaging in order to avoid difficulties in the operation of the slicer and undesirable moisture condensation within the package. The interior crumb temperature should be reduced to a range of 35–40.5 °C before packaging. This prevents moisture condensation, soggy crust and deformation of the wrapped loaf of bread. Cooling also facilitates the redistribution of moisture in the product. The migration of moisture from the centre to the crust softens the crust. The cooling rate depends on air temperature, relative humidity, velocity, flow pattern, bread size and temperature. Cooling bread with a low air temperature can create a hard surface on the crust quickly.

8.2.4 Packaging

After cooling, the bread is sliced and packaged. Bread packaging materials possess certain properties. The bread has to act like a barrier against contaminating agents such as dust, microbes, etc. It is expected to conserve moisture, prevent rapid desiccation and staling, and avoid internal condensation

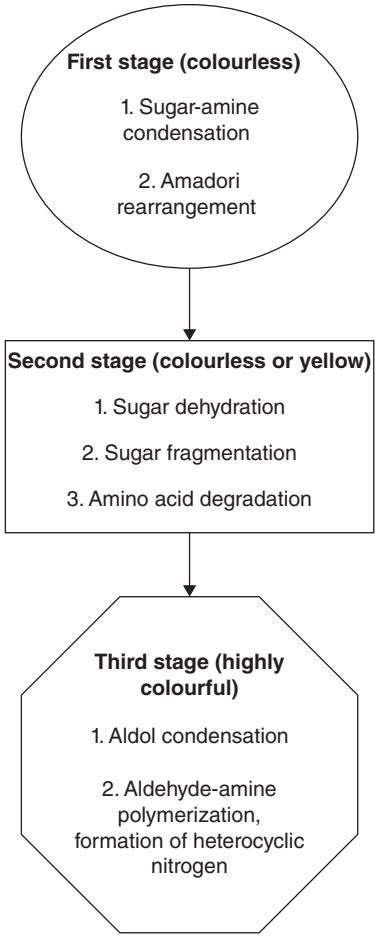

Figure 8.2 Three stages of Maillard reaction

of moisture. Further, packaging provides physical strength against deformation of the product during transportation and storage, and be economical. To improve the microbial safety during packaging and storage, use of modified atmosphere packaging is preferred. Modifying the atmosphere inside a package using ethanol, nitrogen or oxygen is another approach. Not only can it inhibit growth of microorganisms but it can also slow down the staling rate. Waxed paper used for bread and bun packaging is generally made of a base paper made of bleached sulphite pulp with a coating of blend comprising paraffin wax, microcrystalline wax and hot melts. Newer packaging materials like cellophane and polypropylene, which provides good clarity, stiffness, rigidity and good machinability, are being increasingly used. Developments

in the field of bread packaging include certain process modifications in the product to provide a longer shelf-life.

8.3 Biscuit

The term biscuit is derived from the latin word *bis coctus*, which means twice baked. Biscuits are dry, small and thin varieties of cake. Biscuits are small products mainly prepared from flour, sugar, fat and other minor ingredients. Biscuits have a moisture content less than 4% and possess a longer shelf-life. The low moisture content, coupled with the thinness of the products gives them an attractive crisp texture and hard eating characteristics. Once packed, they can be stored for six months or more. Biscuit is a cereal-based product that acts as a predominant source of energy in the human diet.

The low moisture content and low water activity of products in this group mean that they have a long mold-free shelf-life. The organoleptic shelf-life is also long since the product staling and moisture loss are not usually a problem. However, these biscuits are to be protected from absorbing surrounding atmospheric moisture, which can lead to softening of the products and loss of crisp eating characteristics. The second problem is the risk of fat rancidity arising from the combination of a long storage time with low water activity (Manley, 2011).

8.3.1 Classification of biscuits

Biscuits are classified based on dough characteristics as soft dough and hard dough biscuits. Soft doughs lack elasticity and extensibility and these biscuits are crumbly in nature due to the lower amount of added water, the lower mixing period and the higher amount of fat and sugar added. Hard doughs are elastic and extensible. Biscuits are also classified on the basis of taste as sweet biscuits, semi-sweet biscuits and salted varieties. The classification of biscuits is shown in Table 8.2.

Sweet biscuits Sweet biscuits are also known as soft or short dough biscuits having higher sugar and fat contents. They are usually produced using weak flour that forms relatively weak gluten. The desirable moisture content of flour is 12–14% in order to provide proper water absorption (Hui, 2006).

Semi-sweet biscuits These are also known as tea biscuits. They are produced from hard dough and have low sugar and fat contents. The doughs of semi-sweet biscuits are extensible but lack elasticity. In the manufacture of semi-sweet biscuits, the limited gluten development is important, which is achieved by dough aeration.

Table 8.2 Classification of biscuit

Type of biscuit		Sweet biscuit		Semi-sweet	Cracker	
	Sweet moulded	Sweet salted	Sweet wire-cut		Soda cracker	Cream cracker
Sugar (%)	30–40	25–35	45–75	15–20	5–12	5–6
Fat (%)	15–25	12–20	50–60	10–15	15–20	28–30
Moisture content in dough (%)	10–15	15–18	8–12	22	30	30
Moisture content in biscuit (%)	1–2	1–2	1–2	1–2	1–2	1–2
Temperature of dough (°C)	21–22	21–22	21–22	40–42	30–38	30–38
Shaping	Rotary moulding	Rout press	Wire-cut	Sheeting and laminating	Sheeting and laminating	Sheeting and laminating
Baking time (min)	4–5	4–6	8–12	5.5	3	3
Oven band type	Steel	Steel	Steel	Wire	Wire	Wire

Salted varieties Salted varieties include crackers and puffed biscuits. Doughs of salted biscuits are elastic and extensible. These biscuits are produced from either fermented or unfermented hard dough, and have negligible or very low fat and sugar contents. However, the apparent fat content is high due to oil layering or spraying.

8.3.2 Ingredients and their role

The ingredients are selected based on technical, functional, type of biscuits and cost aspects.

Wheat flour Flour is the main ingredient in most biscuits; it contributes to the shape, texture and hardness of biscuits. Usually, flour quality of 70–72% extraction rate and weak to medium strong varieties are preferred. Strong flours can be diluted by adding 10–15% of nonwheat flour like maize, tapioca, rye and barley. Same quality flour is not suitable for making different types of biscuits because dough characteristics are influenced by the quality of gluten proteins. Most biscuits can be made from flour that has a low quantity of protein and gluten that is weak and extensible. Wheat flour having a gluten content of 7–9% is suitable for sweet and soft dough biscuits. The main functions of flour in biscuit making are that it should hold all the ingredients uniformly in the dough, has easy machinability, retains gas during fermentation and baking, and produces a proper structure of the biscuit.

Sugar Sugar is an important ingredient of most biscuits. In addition to sweetness, they influence texture, colour and flavour of biscuits. The amount of sugar used in biscuit making depends on the particle size of sugar; it also influences the spread of biscuits and machining properties of dough to a great extent (Matz and Matz, 1978). Increasing the sugar level generally increases the spreading characteristics but reduces the thickness of biscuits (Vetter *et al.*, 1984; Kissel, Marshall and Yamazaki, 1973; Finney, Yamazaki and Morris, 1950). Sugar has been reported to restrict the development of gluten by competing for water that otherwise may have been absorbed by the gluten (Yamazaki, 1971).

The different physical forms of sugar used in biscuits are granular sugar, castor sugar, pulverized sugar and icing sugar. Sugars used in biscuit making are sucrose, glucose, fructose, malt extract, invert syrup, golden syrup and honey. Reducing sugars such as fructose and glucose impart a desirable golden brown colour to biscuits (Sai Manohar and Hairdas Rao, 1997).

Shortenings Fat is one of the most important ingredients in biscuit manufacture. It improves eating quality by adding flavour to the product and contributes to the structure of biscuits. The key function of fat in biscuits is to

shorten the dough to offer the typical 'melt in the mouth' feature and crumbly texture that are the desirable characteristics of a biscuit (Manley, 2011). Fat acts as a lubricant during mixing of the dough. It also competes with the aqueous phase for the flour surface and disrupts the formation of a gluten network in the dough. Starch swelling and its gelatinization are also reduced at high levels of fat, giving a crisp texture to the biscuit. The type and quantity of fat used have a great influence on the shortening effect, and hence the quality of biscuits. Oil brings about the greatest change in rheological characteristics compared to bakery shortenings and hydrogenated fat, indicating the importance of the solid fat index (Sai Manohar and Hairdas Rao, 1999). Fat contributes to an increase in spreading characteristics and reduction in thickness and weight of biscuits. Emulsified shortenings improve the aeration capacity.

Salt Salt has important functions in biscuits; without salt, the biscuit tastes insipid. It imparts flavour and amplifies and enhances it. Salt tightens the gluten and dough, reduces stickiness and controls fermentation; the most effective concentration of salt is around 1–1.5%. The particle size of salt is important, particularly when used for sprinkling on the surface. High purity vacuum salt is being used in biscuit industries.

Water Water, an important ingredient in biscuit making, is added during the stage of dough preparation, but is driven out during the baking stage. Water hydrates the flour particles and helps in the formation of dough. It has a complex role as it determines the conformational state of biopolymers, affects the nature of interactions between the various constituents and contributes to dough structure (Eliasson and Larsson, 1993). It helps to dissolve chemicals, salt, sugar, water-soluble colours and flavour, and helps to distribute the dissolved materials evenly in the dough. Water helps in the aeration of biscuits to a certain extent by the formation of steam. It is an essential factor in deciding the rheological behaviour of flour doughs (Webb, Russell Eggitt and Coppock, 1970).

Emulsifiers Emulsifiers are substances that allow emulsions to form. Emulsifiers when added to foodstuffs reduce interfacial tension, maintain a uniform dispersion of two or more immiscible substances and aid the wetting of a liquid over a solid. The functions of emulsifiers are to promote emulsion stability, control agglomeration of fat globules stabilize aerated systems to improve texture and shelf-life of products by complex formation with starch components and improve the consistency and crystal structure of fat. The effect of emulsifiers depends on the ratio of fat to water and to the presence of other constituents like starch, protein, fiber and air. Emulsifiers are amphiphilic substances having both lipophilic and hydrophilic portions. The emulsifiers are classified by their hydrophilic–lipophilic balance (HLB). It is the ratio of molecular weights of the hydrophilic portion to the total molecular

weight, and the value ranges between 0 and 20. Most commonly used surfactants in bakery products are sodium stearoyl lactylate (SSL), polysorbate 80 and 60, sucrose monostearate, diacetyl tartaric acid esters of monoglycerides, sorbitan monostearate and propylene glycol monostearate. Most commonly used emulsifiers are mono- and diglycerides. They crystallize in bilayers and produce different lamellar structures. A lamellar mesophase is efficient in promoting the interaction between monoglyceride and amylose, and can provide the antistalling effect. Sodium stearoyl lactylate brings about changes in dough characteristics and the spread ratio of biscuits whereas glycerol monostearate and lecithin improve the quality of biscuits (Sai Manohar and Hairdas Rao, 1999). Lecithin is a naturally occurring emulsifier and has been extensively used in different food formulations. It aids the dispersion of fat in semi-sweet doughs and improves the emulsification during cream-up in short doughs. The usage rate of lecithin is 0.5–1.0%. It is dissolved in fat and added during the creaming stage.

Leavening agents Leavening is defined as a raising action that aerates dough during mixing and baking. Leavening agents used in bakery products help to increase their volume and improve texture. The leavened bakery product becomes lower in density and porous in structure, which can be easily chewed and digested. Leavening of bakery products can be achieved by:

(a) Mechanical method. Air is incorporated by creaming and beating by using proteinatious substances. During the creaming process, air is incorporated into the shortenings, which expands to achieve larger volumes during baking.
(b) Biological method. Yeast is used as a source of biological leavening. However, leavening by yeast is costlier than chemical aeration. In the case of crackers, leavening is done by using yeast.
(c) Chemical method. Some of the commonly used chemical leaveners are baking soda, baking powder and ammonium bicarbonate. Sodium bicarbonate liberates carbon dioxide during baking and helps in leavening. Potassium bicarbonate can replace baking soda to produce sodium-free or low-sodium baked products.

Ammonium bicarbonate is also known as ‘volatile salt’. During baking, it decomposes to form ammonia, carbon dioxide and water vapour. The ammonia gas produced is driven off during baking, which may impart flavours if present in the baked product. Ammonium bicarbonate has an excellent aeration property, but must be used judiciously. Excess amounts can completely break down the structure of the biscuit.

Baking powder is produced by blending water-soluble sodium bicarbonate with one or more acid-reacting ingredients with or without inert fillers such as starch, calcium carbonate or flour. When dissolved in water, the acid and

alkali (sodium bicarbonate) react to emit carbon dioxide. The carbon dioxide thus produced expands the existing bubbles to leaven the moisture. Baking powders are classified into single-acting and double-acting baking powders based on the acid salts present. Single-acting baking powders react at low temperatures, whereas double-acting baking powders react at room temperatures and at higher temperatures cause a further rise during baking (Potter and Hotchkiss, 1995).

Milk and milk products These are generally used in the preparation of sweet and semi-sweet biscuits. Milk products impart a typical flavour and contribute to taste, colour and nutritive value, and improve the texture and tenderness of the biscuit. The protein and reducing sugars (lactose) in the milk products participate in Maillard reactions, which give a golden brown surface coloration to biscuits during baking. Liquid milk is rarely used while skim milk powder, butter and butter oil, cheese and cheese powder, and whey powder are conventionally employed. When milk powder is used, the water requirement increases and improves the flavour of the product. Whey powder contains high amounts of albumin, which has a tenderizing effect on biscuit texture.

Enzymes An enzyme in baking technology improves the quality of bakery products and optimizes the dough properties. Wheat contains many enzymes out of which amylases, proteases and lipooxygenases are of importance in the field of baking. Amylases help in the formation of fermentable sugars and increase the fermentation rate. In biscuit making, the use of amylase is rare except occasionally with yeast fermentation. Proteases are used to modify the gluten quality by acting on the inner peptide linkages of gluten proteins, making the dough softer and extensible (Mathewson, 2000). Sodium *meta*-bisulfite also performs the same function as protease but breaks the chain in a different manner. The biscuit texture obtained by protease is usually more open and tender than that of sodium *meta*-bisulfite. Protease reduces the dough mixing time and reduces viscosity and elasticity, which are the desirable features for the machining of certain biscuit doughs. Proteases added in bakery products are cereal, fungal and bacterial in origin, all of which are inactivated during baking. The use of an oxidation-sensitive protease, such as papain, in combination with an oxidizing enzyme producing an oxidizing agent, can enable biscuit manufacturers to mimic the effect of sulfite in dough; it also improves texture and colour. Lipooxygenase acts on polyunsaturated fatty acids; it is normally used for bleaching of flour pigments and increases mixing tolerance and dough handling characteristics.

8.3.3 Biscuit processing

Biscuit processing method involves several unit operations such as mixing, shaping, baking, cooling and packaging.

Mixing process Mixing of doughs is an important step in the process control of biscuit making. The optimally mixed dough possesses suitable rheological characteristics for further processing in shaping machines. The type and size of the mixer should be considered critically and the preparation of pre-mixes of certain ingredients may improve the technology of mixing. Mixing is done for a minimum period till the dough is formed so as to obtain tender and crisp biscuits. Mixing is to be done in such a way that all the ingredients are well dispersed and the dough is cohesive, enabling easy shaping of biscuits. The nature of dough allows an impression of complex and intricate design on the surface. The consistency of dough is highly important in determining the quality of biscuits. Biscuit doughs are complex as they are made up of a liquid phase that has fat and water and a solid phase that includes starch, protein, sugar and often many other ingredients. Doughs change on standing and rheological properties of hard dough can be modified by the addition of enzymes and sodium *meta*-bisulfite. About 50–100 ppm of *meta*-bisulfite can bring dramatic changes in the dough quality by breaking the disulfide bond of gluten. Sodium *meta*-bisulfite, which is mostly used in semi-sweet hard doughs, reduces the mixing time, baking time, water requirement and final dough temperature. At the end of the mixing period, ingredients absorb water slowly, which results in hardening of the consistency. The different types of mixers that are generally used for biscuit dough preparation are (a) vertical mixers, which include planetary and spindle types, (b) horizontal mixers like the z-blade, gridlap type and high-speed type, (c) reciprocating arm mixers and (d) continuous mixers like the barrel type. Biscuit doughs are mixed by two basic methods, the creaming method and the all-in-one method.

Creaming method This can be done in two or three stages; this method is generally used for the short dough method. The spread of biscuit is greater in the creaming method because of less gluten development, which affects the texture of biscuits. Soft doughs (sweet biscuits) are prepared by this method. The dough thus prepared is processed either by a rotary moulder or a wire-cut machine.

In the two-stage creaming method, the mixing time is 10–15 min. This is done by using a gentle speed of mixer by dissolving as much sugar as possible in the available water, followed by adding milk solids, chemicals and flavours into the mixing bowl. This results in a semi-stiff white cream followed by addition of the flour and is mixed until uniform dispersion of the cream over the flour takes place. The dough formed is pressed into a sheet via a rotary moulder.

In the three-stage method, mixing is done for 10–15 min initially by adding fat, sugar syrup, emulsifiers and part of the water. Then salt, alkaline chemicals and flavours are dissolved in the remaining water and mixed well to obtain a smooth cream. The desired dough consistency is obtained by

adding the flour. This type of mixing encourages the formation of emulsified fat, sugar and water, which leads to less gluten development.

All-in-one method This is a direct and straight-forward method. All the ingredients like salt, leavening chemicals, colour, flavour and water are placed in the mixer. Due to a longer mixing time, gluten development takes place as water is highly accessible to the flour. The formed dough is dense and tough with the dough temperature ranging from 20 to 24 °C; hard dough biscuits are prepared by this method.

Fermentation Fermented doughs are prepared in the case of the cracker. It can be produced by two methods. In the all-in-one mix and fermentation method, all the ingredients including yeast suspensions are mixed together and allow 3–8 h of fermentation. In the two-stage mix and fermentation method, 70% of the flour along with water and yeast suspensions are mixed and fermented for 19 h to form sponge doughs. This is followed by the addition of the rest of the ingredients and allowed to ferment for a shorter period, before being taken for further processing.

Shaping of biscuits After mixing, the dough is formed into pieces by laminating, sheeting and cutting.

Rotary moulders Short dough biscuits are processed using rotary moulders. This consists of a set of rollers consisting of a grooved roller for forcing the dough and a moulding roller in which biscuit shapes are engraved. The advantages of using rotary moulders include (a) it is not necessary to form and support a dough sheet, (b) difficulties of gauging are eliminated and (c) there is no cutter scrap dough.

The dough is forced into moulds, which are the negative shape of the dough pieces complete with pattern and docker holes. The dough is then drawn from the hopper into the nip against the roll.

Wire-cut method Softer doughs, which are rich in fat and sugar, are processed in these machines. Wire-cut machines involve a device that cuts off extruded dough pieces emerging from the die orifice. The cutting part of the device is composed of a blade or a wire drawn through the dough fast by a harp moving back and forth below the orifice. The dough is extruded through dies by rotating rollers and the extruded dough is cut into the desired size with the help of a cutting assembly.

8.3.4 Lamination

This is also known as layering of the dough, which is done by employing laminators. The dough is squeezed between the sheeting rolls and folded, and

finally sent back into the rolls. Thinning of the dough is carried by passing it through two or more thinning cylinders. Lamination of the dough provides a way to repair poor dough sheet, which tends to have been prepared with a simple pair of rolls. It makes the dough more suitable for baking with rolling and folding. The dough is compressed to remove air and inevitably is rested to build the gluten structure. Many large-scale biscuit factories use automatic machines like vertical and horizontal laminators. Semi-sweet biscuits from laminated doughs have excellent textural properties.

Sheeting and cutting Sheeting is done by forcing the dough through sets of polished gauges. Many types of biscuits are made by sheeting the dough from the mixer through pairs of rolls, and then by cutting out the individual pieces from that sheet to make them ready for further processing or baking (Manley, 2011). Sheeting produces a thin film of smooth dough sheet.

The stamp or emboss cutting machine cuts the dough sheets into biscuit shapes by two actions. It swings to and fro and up and down perpendicular to the dough sheet movement. The cutter has biscuit-shaped sheets bolted inside along with decoration and dock pins. The biscuit shapes formed are transferred to an oven band.

Roller cutting is a simple and widely used method. Roller cutters are of two types, one with two rolls and the other with only one roll. In two-roll systems, one roll impresses the sign and the second cuts the dough around the design. The roller cutter with a single roll achieves both dockering and outline cutting with the help of only one roll.

Baking Dough pieces undergo physical and chemical changes inside the oven, which includes the development of rigid porous structures, reduction in moisture content, surface colouration, liberation of gases, conversion of water to steam and starch gelatinization. During the initial stages of baking, moisture present on the surface is lost, which helps in developing crust thickness. Shortenings used in the biscuit melt as soon as their immediate area in the dough reaches the melting, fusion and slip point temperatures of a shortening structure. Water used in dough preparation is converted into steam and carbon dioxide formation; both of these factors contribute to the expansion of the dough pieces, resulting in a volume increase in the baked biscuit. At the mid-stage of baking, the temperature reaches the boiling point of water and hence gluten and other proteins coagulate, which imparts strength to the biscuit structure. This is followed by starch gelatinization to the structure during baking. At the final stage of baking, caramelization of sugar takes place, which produces melanoidins to produce a brown crust. The major noticeable changes during the baking of biscuits are darkening of the surface colour, flavour and aroma development through the Maillard reaction and the considerable loss of water. The conventional final moisture content of the product is 1–3%.

Cooling One of the main reasons for cooling baked products is to prevent condensation after they have been packed. Any condensation within the package may encourage microbial spoilage, particularly mold growth on the surface. The biscuit coming out of the oven is flexible in texture, which is converted into a rigid structure by cooling. Biscuits leaving the oven are at about 100 °C; the speed of cooling is influenced by ambient temperatures and the thickness of the biscuits. During cooling, the interchange of moisture between the product and atmosphere takes place. Slow cooling of biscuits is necessary to avoid the hairline crack formation known as 'checking'. Rapid cooling hardens the biscuit structure. During cooling, the rigid structure results due to solidification of the sugar and fat. There are two systems of cooling: (a) atmospheric multitier conveyer, where the biscuits travel on the canvass web having tiers and are cooled by the surrounding atmosphere, and (b) the forced draft cooling conveyer, where atmospheric cooling is replaced by forced draft cooling. In this case, filtered air is blown against the biscuits coming out of the oven on the cooling conveyer. Biscuits are cooled at a faster rate than the normal time.

Packaging The objective of packaging is to collate the biscuits in groups of suitable size for sale and to protect them so that their crispness, flavour and appearance are preserved as long as possible. The packaging material must form a moisture barrier and resist mechanical damage. Packaging material retards the effects of chemical change by reducing the intensity of light and excluding oxygen. Flexible packaging materials are widely used for unit packing of biscuits. Some of the conventional packing materials used for packing biscuits are wax paper, coated cellophane, polypropylene films and laminates of metalized polyester.

8.4 Cake

The cake-making process has changed in the last few centuries. Marginally, there are a variety of cake products with a broad range of formulations. The main attributes of a cake are structure, texture, moistness, colour (brown crust), high volume and a sweet flavour. Cakes contain higher levels of flour, shortening, sugar, eggs and milk. The modern cake is characterized by a sweet taste, short and tender texture and pleasant flavour (Pyler, 1988). Sponges and cakes represent a more diverse group of products than bread and other fermented products. They do, however, have some unifying characteristics that distinguish them from other baked products. They may be classified as intermediate moisture foods though the total moisture content is lower by about 10–20% of that of bread.

The two basic categories of cakes (foam and shortened) are distinctly different in their preparation and problems. The shortened-style cake (pound

cake, yellow cake, chocolate cake, etc.) has a crumb structure derived from a fat–liquid emulsion that is created during batter processing. Foam-style cakes (angel food, sponge and chiffon) depend on the foaming and aeration properties of eggs for their structure and volume. The quality of a cake is dependent on several factors. The selection of ingredients and the knowledge of their function is the first step towards a quality product.

8.4.1 Classification

Foam-style cakes depend on egg for their structure and volume. This is dependent on the ability of eggs to occlude air and to form stable foams. Angel food cake is a 'true' foam-style cake. This means that the cake is leavened only by air and steam, with no chemical leavening agent. Angel food cake has one of the simplest cake formulas as it calls for only three basic ingredients, egg white, sugar and flour, but may include minor amounts of salt, cream of tartar as an acidifier and flavouring agent. The proportion of sugar in the cake mix is high because no other tenderizer is used; it interferes with gluten development and thus tends to produce a more tender and fragile cake. Sugar also has a stabilizing effect on egg white foam. The air incorporated into the angel food batter serves as the sole leavening agent, so that the role of the developing vapour pressure is critical for the final volume and texture of the final cake.

Sponge cakes do not differ much from angel cake except that both egg yolk and egg whites are used. In some instances, bakers prefer to separate the whites and yolks of the eggs and beat them separately with an appropriate quantity of sugar to attain maximum batter volume. Chiffon cake is a cross between a foam-style cake and a shortened cake. It contains a larger proportion of beaten egg whites to help with leavening along with chemical leavening. It also contains liquid oil in the formulation. Pound cake represents the oldest example of an aerated fat-containing cake. It has no added leavening agent apart from air. Pound cakes have a closer grain and are compacted in character. Cakes also contain some flavouring ingredients.

8.4.2 Ingredients and their role

Ingredients play a major role in creating an acceptable product. Whether alone or together, each ingredient contributes an important quality attribute to the finished cake. Flour, liquid, sugar, leavening agent, egg and fat are mixed in an appropriate proportion to obtain the desirable cake batter yielding a quality product.

Flour Cake flours are normally the milled soft red and white winter wheat varieties. It is pure white in colour and has a very fine and silky soft texture. Cake flour contains a low protein content of about 8% and small particle

sizes compared to an all-purpose flour. It results in less gluten formation, which gives the cake a fine grain, a delicate structure retaining gas and forms a fine foam structure and a soft texture. The flour must yield weak gluten and should not develop a hard texture. Cake flour is treated with chlorine gas, which lowers the pH from about 6.0 to 5.0, bleaches the plant pigments and improves baking quality (Fustier and Gelinas, 1998). Chlorination improves the functional properties of flour components such as lipids, starch, pentosans, proteins and water-soluble substances, as long as their modification is held to a low level. Chlorination increases the water absorption capacity of starch and thus contributes to a firmer and smaller crumb, improves volume and texture, and yields a stable product. This is especially important with the high amount of sugar in the cake formula. Since sugar is a good competitor for water, chlorination aids the starch in this capacity (Conforti and Johnson, 1992).

Shortening Shortening is the primary tenderizing agent in cake and performs three basic functions in cake production: (a) it entraps air during the creaming process, which aids in the leavening of batter and the finished cake, (b) it disrupts the continuity of the gluten and starch structure that makes up the cake crumb by coating the protein and starch particles and (c) it emulsifies large amounts of liquid that contributes to the softness of the cake.

Shortening also imparts moistness; a layered cake made with oil as the shortening gives the impression of a moist crumb rather than a cake from the same formula made with an emulsified plastic shortening (Stauffer, 1998). The hydrogenation process creates a plastic fat that is important to the creation of a cake batter that traps air bubbles and produces a fine-grained and a high-volume cake. The reduction of fat in cake has been investigated, but reducing the amount of fat not only reduces energy (calories) but also cake quality. Various fat substitutes can be used for this purpose; they are categorized as protein or carbohydrate or fat-based ingredients.

Egg Eggs perform multiple functions that affect the structure, volume, tenderness and eating properties of the final product. Eggs are a rich source of protein; they can be whipped into foam, where the proteins are denatured, forming a relatively stable aerated structure that is capable of carrying other ingredients. Egg proteins provide structural support by forming a complex network between the egg protein and flour gluten. On heating, an egg protein network is coagulated, imparting rigidity to the cake crumb. Lipoproteins in egg yolk act as emulsifiers and assist in aeration and foaming (Shepherd and Yoell, 1976). The entrapped air expands when the egg foam is heated, thereby increasing the volume of the foam. Egg yolk also acts as a tenderizing agent as it contains high levels of lipids. The lecithin in the egg yolk acts as an effective emulsifying agent. Egg imparts a relatively mild and distinctive flavour. If a large proportion of egg is used, flavour is transmitted to the cake, so the

need for selecting the eggs for their fresh flavour becomes obvious, that is as shell eggs, as frozen or as separated whites and yolks. The colour contributed by eggs to cake is of considerable significance. Egg white may contribute liquid to a product and thus serve as a toughener due to its partial contribution to gelatinization of starch and gluten development. The stability of the egg white foam is largely influenced by the pH of the egg white. The lower the pH, the more stable the foam. Sugar is added to egg white foam to form a meringue. The addition of sugar stabilizes the foam. When beating egg whites, the whites go through four stages: foamy, soft peak, stiff peak and dry. In addition, eggs increase the food value of the product and impart a better colour and appearance to the finished products. Whole eggs are beaten into foam for some cakes.

Sweetener The principal sweetener used in cake is sucrose. Sugar serves multifunctional roles in cake making. One important function is that it increases the volume of cakes by the incorporation of air into the fat during creaming. During baking, sugar raises the temperature at which gelatinization and coagulation occur, which gives the gluten more time to stretch, thereby further increasing the volume of the baked product and contributing to a finer and more even texture (Bean and Yamazaki, 1978). The hydroscopic or water-retaining nature of sugar increases the moistness of the baked cakes. This is evident when brown sugar is used in the formulation. Sugar competes for water with starch and protein. Further, sugar helps to brown the crust through caramelization and Maillard browning.

Emulsifiers Emulsifiers promote the incorporation of air in a cake batter in the form of fine bubbles and disperse the shortening into smaller particles. Emulsifiers act uniquely at the boundaries of the oil–water interface to maintain a homogeneous mixture. An emulsified shortening containing monoglyceride is frequently used in cakes (Stauffer, 1998). Other emulsifiers such as polysorbate 60, sorbitan monostearate, and mono- and diglycerides give good results.

Leavening agents The initial leavening agent incorporated into cake batter is air. Carbon dioxide is incorporated either by a biological source (yeast) or by chemical sources (baking powder and baking soda). Air is incorporated by the simple action of sifting together the flour and other dry ingredients (salt, baking powder and/or baking soda). Another method that incorporates air is 'creaming' of fat and sugar. During heating, the air is released and the product rises. Beating egg whites is another example of incorporating air into the batter. Chemical leavening involves baking powder and baking soda (sodium bicarbonate). When either of these compounds comes in contact with a liquid and heat, CO_2 is produced and the product rises. The two types of baking

powder used in bakery are fast- or single-acting baking powder and slow- or double-acting baking powder.

Liquid Whether the liquid in the formula is water, milk or buttermilk, it performs several functions in the recipe. Water acts as a plasticizer. The amount and type of dissolved minerals and organic substances present in water can affect the flavour and colour. Water helps in the gelatinization of starch in the flour, dissolves the ingredients, especially sugar, during mixing and baking of cake batter. It also aids in the release of carbon dioxide from either baking powder or baking soda. Water also produces steam and therefore, along with carbon dioxide, aids in leavening. Usually milk is a popular liquid for use in cake batter. In addition to contributing water, milk gives richness to the cake, better bloom and crust colour, helps in the incorporation of more and larger air cells and prevents curdling during creaming. The lactose in the milk participates in the Maillard reaction, resulting in a brown crust. Water in milk also acts as a moistener to help in the development of gluten.

Salt Salt performs three important functions in cakes: (a) adjustment of sweetness, (b) extract flavour from other ingredients in the cake and (c) lowers the caramelization temperature to aid in obtaining the desirable crust colour.

Hydrocolloids Hydrocolloids in the food industry are used to improve texture, shelf-life, moisture-retention capacity, cake volume and crumb grain, and slow down the retrogradation of starch. Hydrocolloids modify the amylograph characteristics of starch by affecting the baking process and final quality of cakes (Rosell, Rojas and Benedito de Barber, 2001b; Rojas, Rosell and Benedito de Barber, 1999; Christianson *et al.*, 1981).

8.4.3 Formula balance

The most important criteria for obtaining a good quality cake are formula balancing. Variation in one of the ingredients requires counter balancing with other ingredients. The rules of formula balance are that (a) the weight of sugar is equal to the weight of the flour, (b) the weight of shortening is equal to the weight of the eggs and (c) the liquid ingredients like milk and eggs are equal to the weight of the flour or sugar. Too much fat or sugar weakens the structure, excess baking powder results in collapse of the cake in the centre and an excess of liquid toughens the structure. The introduction of emulsified shortenings enables high amounts of water to be used because of the flowing nature of batter, and thus the capacity to retain gas falls (Pyler, 1988).

The formula balance for a batter-type cake can be divided into high-ratio and low-ratio cakes. The formula balance for low-ratio and high-ratio cakes are:

(a) Low-ratio cakes. Sugar should not be more than flour, total liquid must be equal to the liquid in egg and milk, total liquid must be equal to sugar and shortening should not exceed the amount of egg.
(b) High-ratio cakes. Sugar must be greater than flour, total liquid must be equal to the liquid in egg and milk, total liquids must be more than sugars and egg can be equal to or greater than shortening.

Foam-type cakes The weight of the sugar should be equal to the weight of the egg. The weight of the flour should be approximately one-third the weight of the sugar. Foam-type cakes are classified into angel food cake and sponge cake. Angel food cake is based on a simple formula of one part of flour to three parts each of egg whites and sugar. The sponge cake formula includes the amount of sugar that slightly exceeds or equals the amount of the whole egg. Weights of the liquid in the whole egg and milk or water should exceed the weight of the sugar by the ratio of 1.25:1. The weight of sugar and egg should exceed that of flour.

Pound cake Formula balances for pound cakes are that the weight of egg should be equal to or greater than fat, the weight of sugar should be equal or slightly more than that of the flour and the combined weight of liquid ingredients should equal the weight of the flour or sugar, whichever is greater.

8.4.4 Mixing process

The procedure of mixing depends on the nature of the cake being produced. The primary purpose of cake mixing is to bring about a complete and uniform dispersion and to incorporate air into the mix. The incorporation of air takes place during a period of rapid incorporation in the form of large bubbles and a stabilizing period when the bubbles are reduced in size (Hui, 2006) and there is mutual emulsification of ingredients.

Mixing methods for batter type cakes are (a) sugar batter or creaming method, (b) flour batter or blending method, (c) single-stage method and (d) emulsion method. In the sugar batter or creaming method, large volumes of air are incorporated in the form of minuscule cells in the fat phase of the batter, coating of flour by fat and sugar, which delays their respective hydration and solubilization characteristics, and the near absence of flour gluten development. Initially, shortenings and granulated sugar along with

dry ingredients are mixed at a medium speed until they are aerated. Mixing is completed by the addition of egg, milk and flour. Mixing time is important; overmixing causes a loss of air and produces a heavy cake. The total mixing time for the creaming method is between 15 and 20 min; the initial creaming stage takes 8–10 min, the second stage with the incorporation of the egg is 5–6 min and the final stage of milk and flour addition is 5–6 min. If the milk is added too quickly, it causes curdling of the batter and inversion of the emulsion to a water-in-oil emulsion (Pyler, 1988).

In the flour batter or blending method, shortening and flour are creamed; simultaneously, eggs and sugar are whipped at a medium speed to obtain a semi-firm foam in separate bowls. These steps require about 10 min. The sugar–egg foam is combined with creamed flour– shortening mixture followed by gradual addition of milk. This method produces fine grain and uniform texture in the cake. The disadvantage of this method includes the lesser incorporation of air, which results in a low product volume and pronounced development of gluten, which gives toughness to the finished cake. In the single-stage method, all the ingredients are mixed at one time to produce a homogeneous mass. Baking powder is incorporated at the final mixing stage. The total mixing time is 8 to 10 min. The emulsion method is suited for large-volume cake mixers. Sugar and shortenings are creamed together, followed by the addition of milk in several portions, with continuous beating until a light and fluffy mass is obtained. Finally, flour and eggs are added. The total mixing period is 12 to 15 min.

The mixing method employed for a foam-type cake incorporates air to form stable foams, which affect the structure and volume of the product. In addition, as beating continues, the foam loses its sheen and reaches its maximum volume and stiffness. Foam-type cakes are mixed by the continuous batter mixing method. It can be performed by using two basic types of continuous batter mixers, such as the compact rotor stator mixing chamber type and the tubular scraped or swept surface mixer.

8.4.5 Baking

Baking is probably the most important factor governing the quality of the final cake. Once the cake batter is mixed, it should be deposited into cake pans and conveyed to the oven with a minimum loss of time so that the inevitable escape of carbon dioxide from the batter and coarsening of the cell structure are prevented. The optimum baking conditions for a cake are determined by factors such as the sweetener level, amount of milk, pan size and fluidity of the batter. The baking time is inversely proportional to the baking temperature.

Cake pans are available in a variety of shapes and forms, ranging from flat sheet pans to cupcake moulds. Crumb formation is partially dependent on

the degree of heating that occurs when the cake batter is first placed in the oven and rapid heat absorption plays a major role. On the other hand, shiny surfaces reflect heat, which causes the cake to take longer to bake and results in a coarser grain having a lower volume (Brown, 2000).

Cake ingredients must be modified at altitudes higher than 1000 m. The lower atmospheric pressure at higher elevations reduces the need for baking powder or baking soda. Also, water evaporates more quickly and the concentration of sugar increases. Other factors that deserve consideration at higher altitudes include: (a) batter and foam-type cakes should be mixed at higher specific gravities than usual at sea level, (b) the cake pans must be coated heavily to prevent sticking during depanning the product, where the use of silicon-coated pans is helpful to minimize the problem of sticking, and (c) egg whites should be used to adjust the amount of eggs to the required level rather than egg yolks because the batter lacks sufficient moisture and protein, which provide the necessary batter stabilizing effect. Structural strength can be improved by adding 1 to 2 tablespoons of cake flour, increasing the amount of liquid or reducing baking powder and/or baking soda and sugar quantities. An increase of the baking temperature by 6–8 °C increases the rate at which the cake sets by speeding the coagulation of protein and the gelatinization of starch. Cakes baked at high altitudes show low volumes but tend to exhibit greater crumb tenderness (Pyler, 1988).

8.4.6 Post-baking operations

Cooling is a critical stage in cake production as it affects the texture and appearance of the product. Cakes have a high moisture content and tend to dry out rapidly under normal storage conditions. The staling of cakes is caused by two major factors: the first is the movement of moisture through the cake as the dry cake crust attracts moisture and the second is due to changes in the amylopectin fractions of starch. Hence, the packaging material should be selected in such a way that it should prevent moisture movement, microbial growth and loss of aroma and provide physical protection against crushing. Some of the conventional packaging materials include grease proof, glassine paper and cellophane. Newer packaging materials for cakes include thermoformed plastic such as polystyrene, polypropylene (PP) and polyamide-11 film, with vacuum packaging and subsequent IR heat sterilization.

8.5 Machinery

Several machineries like the mixer, moulder, shaping system, baking oven, cooling system and size reduction unit are common in baking industries.

8.5.1 Mixer

Mixing in its most general form can be defined as an operation by which two or more materials having some distinguishable characteristic are brought together through the application of an external force into a closer relationship to one another such that the average distance between the particles of one material and the particles of other material becomes less. The mixing of ingredients covers a number of distinct functions like (a) blending of ingredients to form a uniform mass, (b) dispersing the solid in a liquid or the liquid in a liquid, (c) kneading the mass to impart the development of gluten from flour proteins that have been hydrated at an earlier stage of mixing and (d) aeration of a mass to give a lower density.

Blending or mixing can be achieved by many different types of equipment but they all rely on one of the following types of action or principles of operation: (a) devices using blades, paddles, ribbons, etc., to push a portion of the mix through another portion, (b) devices relying on the elevation and dropping of all or a portion of mass so that the random rebounding of individual particles during the gravitational stage results in mixing and (c) devices creating turbulent movement by injection of currents of a liquid or gas (Matz, 1972). The type of mixers falls into two basic categories: horizontal mixers and vertical mixers.

Horizontal mixers These mixers can be used for a wide variety of mixtures having consistencies from thin batters to extremely tough or dry doughs. More gluten development uses this kind of mixer. In general, all of the horizontal mixers suitable for mixing the usual types of bakery products include a common horizontal mixing bowl, U-shaped in cross-section, mounted on a heavy rigid frame enclosing the dry motor and transmission. Discharging the dough can be done by two methods: (a) the bowl can be tilted by means of a gear motor or a hydraulic mechanism up to an angle of 90 or 140°, which helps in easy ejection of the finished dough, and (b) the bowl is anchored firmly to a mixer frame and removal of the finished dough is accomplished by lowering the bowl door and by slow movement of the mixer arms. Horizontal mixers basically consist of a sturdy frame of cast iron, tubular steel or channel steel, whose upper portion houses the mixing bowl. Some of the mixers consist of a single agitator shaft with three or more mixer bars. Mixing and development of doughs in a horizontal mixer are performed by the rolling, kneading and stretching actions imparted to the dough by cylindrical mixer bars.

Vertical mixers These mixers can perform highly versatile mixing operations and are applied to perform diverse tasks like whipping of foams, aerating of batters and kneading of stiff doughs. A unifying feature of a vertical mixer is the use of movable bowls or troughs. They contain one or more beater shafts, which may be stationary or move in a planetary design. The planetary mixers

are capable of mixing batters and some doughs but most often they are used for adjuncts such as icings. The movement of an agitator is planetary, which revolves around its own vertical axis at a high speed and the axis also moves in circles and rotates around the bowl. The bowls in the planetary mixers are of different sizes, which can be raised or lowered by an auxiliary motor. Agitators are available in different designs like the dough hook, which contains a single curved arm to aid gluten development by a stretching and kneading action. The wire whip contains a set of wires that is wide at the top and pointed at the bottom; it provides the maximum incorporation of air and a bubble dividing action during mixing. Batter beaters having two or four wings are shaped to fit inside the bowl (Matz, 1972).

Cookie and cracker doughs are frequently mixed in spindle mixers. They have a special mobile trough used for fermenting sponges. The action imparted by the blades is tearing, cutting and churning, which are not conducive to dough development. Reciprocating agitator mixers contain a pair of agitator arms that travel through intersecting elliptical paths in a shallow and slowly revolving bowl. These mixers are useful in mixing temperature-sensitive doughs when an intensive blending action is not required. Dough output for vertical mixers per unit time is relatively small as compared to horizontal mixers.

8.5.2 Oven

The oven is the most conspicuous and characteristic piece of equipment in the bakery. It has an important influence on product quality but cannot compensate for errors committed earlier in the processing sequence. The oven is frequently referred to as the 'heart' of the bakery as it performs the ultimate task in converting raw dough into bread, cakes, cookies, etc. The basic types of oven in current use are the reel ovens, single-lap tray ovens, double-lap tray ovens, tunnel band ovens and the most recently introduced oven, conveyorized ovens.

Reel ovens These ovens consist of a reel structure that revolves vertically around a horizontal axis within the baking chamber and supports the baking trays in Ferris wheel fashion. Compared to most other types of ovens, the fuel consumption is high in relation to their production capacity. It is difficult to attain a uniform heat distribution and temperature control due to the presence of reels. These ovens are normally heated by direct firing where electricity or gas is used as the energy source. Heating elements are centrally positioned across the floor of the baking chamber.

Travelling tray ovens The reels here are replaced by two parallel endless chains that carry trays through the baking chamber, with the third chain acting to stabilize the horizontal position of the trays during the baking cycle.

Travelling tray ovens may be single lap or double lap. In single-lap ovens, the trays travel back and forth. A double-lap tray oven travels through four heat zones rather than two as in the single-lap oven.

Tunnel ovens These ovens are long, contain a low baking chamber through which a motor-driven conveyer, carrying the baking hearth, passes in a straight line. In the case of tunnel ovens, the optimum steam conditions are simply and effectively established. It is most suitable for baking of cookies and crackers. Solid steel bands are more suitable for fluid batter products, which inhibit the escape of steam and may cause undesirable cavities in the bottom of crackers (Pyler, 1988).

Conveyorized ovens These ovens have an integrated continuous proofing and baking system. They contain continuous grid-type pan conveyers that carry the pan dough products through both the final proofer and the oven for the required period of time and without interrupting the transfer process. It contains an endless conveyer that is arranged in either ascending or descending spirals or tiers about the periphery. The pan product travels the same path and hence a high degree of uniformity of bake is achieved.

Rack ovens These consist of a vertical baking chamber that is wheeled and a special rack carrying as many as a hundred trays of products. The rack rests on a turned table that rotates during the baking phase and exposes the product to uniform convection baking.

Electronic ovens Electric ovens use electric power for providing convection heating to commercial bakery ovens. High-frequency electromagnetic microwaves are also used. They pass through the product with moderate uniformity, so heating takes place evenly throughout the product. Microwave heating is an effective process for rapid defrosting of frozen fruits, eggs and frozen bakery products and for inhibition of mold growth in packaged sliced bread and other bakery products (Pyler, 1988).

8.6 Conclusions

Bakery products are quite popular among all age groups across the globe and the demand for quality products is increasing world over. Among bakery products, bread, biscuits and cakes find their everyday use. The unit operations for production of different bakery products include a selection of appropriate raw materials, formulation and scaling, mixing, fermentation, shaping, baking, cooling and packaging. Baking is the most unique unit operation in a bakery wherein the intermediate dough or batter is converted to a rigid structure with desirable product characteristics. During baking, heat and mass transfer bring

in physical and chemical changes in the bakery products. In recent years, many alternative baking technologies have been developed, such as jet impingement ovens, microwave ovens and hybrid ovens. More research studies are needed on the design of various types of ovens and on the physicochemical changes that take place during baking in these ovens to improve the process and product quality.

References

Bean, M.M. and Yamazaki, W.T. (1978) Wheat starch gelatinization. 1. Sucrose, microscopy and viscosity effects. *Cereal Chemistry*, **55**, 936–944.

Brown, A. (2000) Cakes and cookies. In: *Understanding Food: Principles and Preparation*, Chapter 25, Thomson Learning, Wadsworth, Australia, pp. 416–989.

Cauvain, S.P. and Young, L.S. (2006) *BakeTran, High Wycombe*, Bucks. In: *Bakery Science and Technology*, Blackwell Publishing, England, pp. 72–98.

Christianson, D.D., Hodge, J.E., Osborne,D. and Detroy, R.W. (1981) Gelatinization of wheat-starch as modified by xanthan gum, guar gum, and cellulose gum. *Cereal Chemistry*, **58**, 513–517.

Conforti, F.D., Johnson, J.M. (1992) Use of the Farinograph in predicting baking quality of unchlorinated and chlorinated flours. *Journal of Food Quality*, **15**, 333–347.

de Boer, L., Heermans, C. and Meima, R. (2005) Reduction of acrylamide formation in bakery products using *Aspergillus niger aspargine*. In: *Using Cereal Science and Technology for the Benefit of Consumers* (Cauvain, S.P., Salmon, S.E. and Young, L.S., eds), Woodhead Publishing, Cambridge, England, pp. 401–405.

Eliasson, A.C. and Larsson, K. (1993) *Cereal in Bread Making: A Molecular Colloidal Approach*, Chapter 6, Marcel Dekker, New York, USA, pp. 261–266.

Finney, K.F., Yamazaki, W.T. and Morris, V.H. (1950) Effects of varying quantities of sugar, shortening and ammonium bicarbonate on the spreading and the top grain of sugar snap cookies. *Cereal Chemistry*, **27**, 30–41.

Fustier, P. and Gelinas, P. (1998) Combining flour heating and chlorination to improve cake texture. *Cereal Chemistry*, **75**, 568–570.

Hui, Y.H. (2006) *Bakery Products Science and Technology*, Blackwell Publishing, USA, pp. 301–314, 337–356, 411–416..

Kissel, L.T., Marshall, B.D. and Yamazaki, W.T. (1973) Effect of variability in sugar granulation on the evaluation of flour cookie quality. *Cereal Chemistry*, **50**, 255–264.

Lane, M.J. and Nursten, H.E. (1983) The variety of odors produced in Maillard model systems and how they are influenced by reaction conditions. In: *The Maillard Reaction in Foods and Nutrition* (Waller, G.R. and Feather, M.S., eds), ACS Symposium Series 215, Chapter 9. American Chemical Society, Washington, DC, USA, pp. 141–158.

Manley, D. (2011) *Manley's Technology of Biscuits, Crackers and Cookies*, 4th edn. Woodhead Publishing Limited, England, pp. 160–118.

Marston, P.E. and Wannan, T.L. (1976) Bread baking: the transformation from dough to bread. *Bakers Digest*, **50**(4), 24–28, 49.

Mathewson, P.R. (2000) Enzymatic activity during bread baking. *Cereal Foods World*, **45**, 98–101.

Matz, S.A. (1960) *Bakery Technology and Engineering*. The AVI Publishing Company, Westport, Connecticut, USA, pp. 35–188.

Matz, S.A. (1972) *Bakery Technology and Engineering*, 2nd edn.The AVI Publishing Company, Westport, Connecticut, USA, pp. 277–410.

Matz, S.A. and Matz, T.D. (1978) *Cookie and Cracker Technology*, 2nd edn. The AVI Publishing Company, Westport, Connecticut, USA, pp. 36–39.

Pateras, I.M.C. (1998) Bread spoilage and staling. In: *Technology of Breadmaking* (Cauvain, S.P. and Young, L.S., eds). Blackie Academic and Professional, London, England, pp. 240–261.

Pomeranz, Y., Meyer, D. and Seibel, W. (1984) Wheat, wheatrye, and rye dough and bread studied by scanning electron microscopy. *Cereal Chemistry*, **61**, 53–59.

Potter, N.N. and Hotchkiss, J.H. (1995) *Food Science* (Jain, S.K., ed.), 5th edn. CBS Publishers and Distributors, Noida, India, pp. 381–407.

Prouty, W.W. (1965) Guidelines for a quality control program 11: process control. *Bakers Digest*, **39**(4), 69–73.

Pyler, E.J. (1988) *Baking Science and Technology*, 3rd edn, vol. 2. Sosland Publishing Company, Kansas City, Missouri, USA, pp. 911–1002, 1184–1199.

Rask, C. (1989) Thermal properties of dough and bakery products: a review of published data. *Journal of Food Engineering*, **9**, 167–193.

Rojas, F., Rosell, C.M. and Benedito de Barber, C. (1999) Pasting properties of different wheat flour-hydrocolloids systems. *Food Hydrocolloids*, **13**, 27–33.

Rosell, C.M., Rojas, F. and Benedito de Barber, C. (2001a) Influence of hydrocolloids on dough rheology and bread quality. *Food Hydrocolloids*, **15**, 75–81.

Rosell, C.M., Rojas, F. and Benedito de Barber, C. (2001b) Combined effect of different antistaling agents on the pasting properties of wheat flour. *Food Research Technology*, **212**, 473–476.

Sablani, S.S., Marcotte, M., Baik, O.D. and Castaigne, F. (1998) Modeling of simultaneous heat and water transport in the baking process: a review. *Lebensmittel Wissenschaft und Technologie*, **31**, 201–209.

Sai Manohar, R. and Haridas Rao, P. (1997) Effect of sugars on the rheological characteristics of biscuit dough and quality of biscuits. *Journal of Science and Food Agriculture*, **75**, 383–390.

Sai Manohar, R. and Haridas Rao, P. (1999) Effect of emulsifiers, fat level and type on the rheological characteristics of biscuit dough and quality of biscuits. *Journal of Science and Food Agriculture*, **79**, 1223–1231.

Sansstedt, R.M. (1961) The function of starch in the baking of bread. *Bakers Digest*, **35**(3), 36–44.

Shakuntala, M. and Shadaksharaswamy, N.M. (2008) *Foods – Facts and Principles*, 3rd edn. New Age International Limited, Ansari Road, Daryaganj, New Delhi, India, pp. 231–251.

Shepherd, I.S. and Yoell, R.W. (1976) Cake emulsions. In: *Food Emulsions* (Friberg, S., ed.), Chapter 5. Marcel Dekker, New York, USA, pp. 215–276.

Stampfl, L. and Nersten, B. (1995) Emulsifiers in bread making. *Food Chemistry*, **52**, 353–360.

Stauffer, C.E. (1998) Fats and oils in bakery products. *Cereal Foods World*, **43**(3), 120–126.

Vetter, J.L., Bright, H., Utt, M. and McMaster, G. (1984) Cookie formulating: sugar, mixing affects specific gravity, spread. *Bakers Digest*, **58**, 6–9.

Webb, T., Russell Eggitt, P.W. and Coppock, J.B.M. (1970) A rheological investigation of the role of water in wheat flour doughs. *Journal of Food Technology*, **5**, 333–343.

Williams, T. and Pullen, G. (1998) Functional ingredients. In: *Technology of Breadmaking* (Cauvain, S.P. and Young, L.S., eds). Blackie Academic and Professional, London, England, pp. 45–80.

Yamazaki, W.T. (1971) Soft wheat products. In: *Wheat Chemistry and Technology* (Pomeranz, Y., ed.). American Association of Cereal Chemists, St Paul, Minnesota,, USA, pp. 749–753.

Yasunaga, T., Bushuk, W. and Irvine, G.N. (1968) Gelatinization of starch during bread-baking. *Cereal Chemistry*, **45**, 269–273.

Zehentbauer, G. and Grosch, W. (1998) Crust aroma of baguettes 11. Dependence of the contaminations of key odorants on yeast level and dough processing. *Journal of Cereal Science*, **28**, 93–96.

9
Frying of Foods

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9.1 Introduction

Frying is one of the oldest unit operations used not only in industry but also at home. Deep-frying is a common multifunctional unit operation for fast drying, texturing and cooking of foods. Intense heat and mass transfer achieved during deep frying led to innovative applications for food materials. As a result of frying, the piece of food emerges sterile and dry in the surface, with a relatively long shelf-life. High temperatures of frying (e.g. >120 °C) eliminate microorganisms from the food piece (favouring its microbiological safety). At the same time, these temperatures favour the reactions between some substances naturally present in the raw food (e.g. reducing sugars and asparagine), inducing the formation of some toxic compounds such as acrylamide and furan, and, in this way, affect negatively the chemical safety of some starchy fried foods (Pedreschi, 2012). During frying, fats and oils can reach much higher temperatures than water at normal atmospheric pressure. Therefore the food sample is cooked quickly and acquires the desirable and attractive sensorial attributes. If the frying time is too long, the surface of the food could even be carbonized while some sugars become caramelized. Depending on the food microstructure, the oil will penetrate the fried food to varying degrees, contributing to lubricity and attractive flavour, as well as to add calories. The quality of the fried product depends not only on the frying conditions but also on the type of oils and microstructure of foods (Pedreschi, 2012). On the other hand, the high

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amount of oil retained inside the fried products is incompatible with current health trends (Moreno, Brown and Bouchon, 2010).

Deep-fat frying is a widely used food process that consists basically of immersion of food samples in hot oil. Deep-fat frying is a rather complex process comprising simultaneous heat and mass transfer with chemical reactions and textural changes taking place. The piece of food in contact with the hot oil heats up and water is lost while the oil content increases; the reaction between reducing sugars and amino acids leads to browning and changes of texture, with softening at the beginning of frying and hardening of the external layers of samples with longer frying times (Pedreschi, 2012). Additionally, deep-fat frying processes could take place at atmospheric pressure (conventional frying) or at pressures below or above atmospheric pressure (vacuum or pressure frying). Commercial atmospheric deep-fat frying is usually performed at high temperatures under atmospheric pressure. Oil deterioration and artefacts produced due to the prolonged and continued use of the oil might have an adverse health effect. Pressure frying is another way of deep-fat frying in which food is fried in a closed system under pressure. It increases the boiling point of the frying oil in the food and therefore shortens the frying time. In vacuum frying, the food is heated under reduced pressure in a closed system that lowers the boiling points of frying oil and the moisture in the food (Garayo and Moreira, 2002). This method allows better retention of the natural colour and flavour as the result of lower applications of heat and oxygen, which are the two essential elements in the oxidation process.

The final oil and moisture content of the fried food will depend on its microstructure, chemical composition, geometrical shapes and process conditions (Hindra and Baik, 2006; Saguy and Danna, 2003). Moisture content is an important property in fried food product quality. Both the oil content and the moisture content are critical parameters that determine the quality and stability of fried products. It is desirable that the moisture and oil contents in fried foods are expressed on a dry basis (free of oil) since both the oil and water contents of the sample being fried change during the process (Moreira, Castell-Perez and Barrufet, 1999).

According to the food microstructure, geometrical shape and frying process conditions, two principal kinds of fried products could be formed after frying (Pedreschi, 2012; Pedreschi *et al.*, 2001). These are (a) thin fried slices (chips) characterized for being a dehydrated and crispy region where oil is located and some toxic compounds such as acrylamide could be distributed inside it, and (b) a composite structure formed by two regions such as an external dehydrated and crispy region, where oil is located and where some toxic compounds such as acrylamide could be distributed, and a humid and cooked core free of oil and acrylamide, and other toxic compounds formed in the surface, which could not migrate into the core.

For the last 15 years, the kinetic studies of quality changes during frying have been more focused on potato products. There were several other

products, such as meat and gluten balls, tortilla and cassava chips, doughnut, tofu and corn starch patty. The study and modelling of the kinetics of the changes in some important physical properties in potatoes during frying have been reported (Moyano and Pedreschi, 2006; Pedreschi *et al.*, 2005; Pedreschi and Moyano, 2005; Pedreschi, Aguilera and Pyle, 2001). However, there are studies on other products such as meat balls, tofu, tortilla chips, chicken nuggets, pork meat and doughnuts (Ngadi, Li and Oluka, 2007; Sosa-Morales, Orzuna-Espíritu and Vélez-Ruiz, 2006; Baik and Mittal, 2003; Vélez-Ruiz and Sosa-Morales, 2003).

9.2 Frying as a unit operation

Immersion frying involves simultaneous heat and mass transfer. Deep-fat frying is a process of heating and dehydration in which the sample is submerged in the oil, which transfers heat very fast into the food. Moisture present in the food is vaporized and forces its way to the food surface. The outside of the food, in addition to being browned by the heat of the oil, is puffed and crisped by this rapid moisture loss. Departing steam increases convection of the oil as it reaches to the surface of the fryer; this convection and the high oil temperature cause uniform and rapid cooking that is characteristic of frying (Pedreschi, 2012). The deep-fat frying process is performed by immersing raw materials in a large volume of hot oil where heat is transferred from the oil to the material via convection at the outer surface and conduction through the solid material. Deep-fat frying, which is also defined as a violent process of drying, may be broken into four stages: (1) initial heating, (2) surface boiling, (3) falling rate, and (4) bubble end point (Farkas, Singh and Rumsey, 1996). The diagrammatical setup of this process is shown in Figure 9.1. The

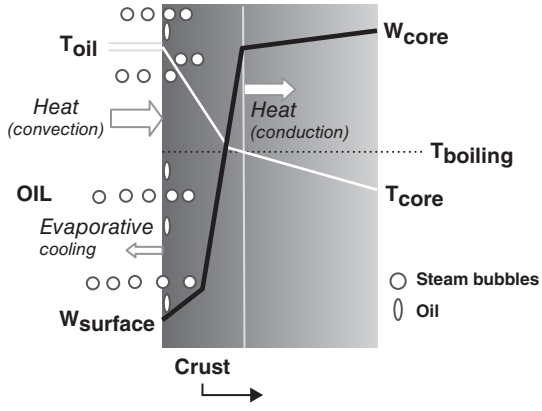

Figure 9.1 Scheme of heat and mass transfer during frying and the prevailing temperatures and moisture profiles (source: Miranda and Agukera 2006. Reproduced with permission of Taylor & Francis Ltd)

high temperatures in deep-fat frying processes (around 160 and 190 °C) cause water evaporation, which is transferred from the food towards the surrounding oil, whereas oil is absorbed by the food-replacing part of the released water (Mellema, 2003).

Initial heating is described as the immersion of a raw material into hot oil and is characterized by the absence of water vaporization. During this stage, heat is transferred from the oil into the food by free convection and through the food by conduction. Surface boiling is characterized by the sudden loss of free moisture at the surface, increased surface heat transfer rate and inception of crust formation (surface drying). The evaporation will also lead to shrinkage and development of surface porosity and roughness. Especially explosive evaporation can lead to the formation of large pores. Water deep inside the food will become heated and will be cooked. The falling rate stage parallels that of drying, in which there is continued thickening of the crust region, a decreased heat transfer rate and a steady decrease in the rate of vapour mass transfer from the food sample. As the food is fried for a longer period of time, the moisture content in the crust slowly diminishes, thereby reducing the amount of steam bubbles leaving the surface. The surface temperature can rise above the boiling temperature of water. Several physicochemical changes such as starch retrogradation, Maillard reactions, glass transitions and acrylamide formation take place (Mellema, 2003). Finally, the bubble end point is characterized by the apparent cessation of moisture loss from the food during frying.

This complex unit operation also involves significant microstructural changes; in fact, most of the desirable characteristics of fried foods are derived from the formation of a composite structure: a dry, porous, crispy and oily outer layer or crust and a moist cooked interior or core are formed during the process (Bouchon *et al.*, 2001). Large pieces of food like French fries or meat balls do not show the temperature of the food core above 100 °C. For thin slices, the core temperatures will be higher. Apart from water vapour other compounds also go from the food to the fat. This, combined with long-lasting high temperatures, leads to the degradation of the frying fat (Mellema, 2003). Some undesirable effects derived from the long-lasting high temperatures involved in the atmospheric deep-fat frying process and exposure to oxygen are the degradation of important nutritional compounds and the generation of toxic molecules in the foodstuff or the frying oil itself.

It is worth noting that the final quality of the fried product is particularly oriented by the dynamic and cumulative contributions of heat transfer that affect respectively: (a) the dynamics of the vaporization kinetics and the intensity of internal mechanical stresses, and (b) the amount of water removed, the fraction and repartition of voids (e.g. pores filled with steam) and the hydrothermal history of the hygroscopic solid matrix (Raoult-Wack *et al.*, 2000). These contributions thus lead to different porous structures,

reaction rates (e.g. browning and enzyme/microorganism inactivation) and oil uptake according to the initial properties of the raw materials, frying conditions and coupled phenomena occurring inside the material during frying and cooling.

Over the last two decades, reducing the oil content of fried products has been of great appeal. Hence, the consumer trend is towards less greasy and healthier products (Ziaiifar, Courtois and Trystram, 2010). The mechanism of oil uptake has been studied primarily to understand the phenomenon and to develop the know-how and means to reduce the final oil content (Danna and Saguy, 2006). However, in spite of all these efforts, fried food products still possess significant amounts of fat, exceeding 30% in some cases (Table 9.1). Oil uptake is mainly a surface phenomenon, as confirmed by many experimental observations (Baumann and Escher, 1995). Concerning the time (moment) of oil absorption, it has been shown that oil does not enter the product to a great extent during frying, but is drawn from the oil film on the product when it is removed from the oil bath (Ziaiifar *et al.*, 2008; Pedreschi *et al.*, 2008; Bouchon, Aguilera and Pyle, 2003). Finally, not only the total amount of oil absorbed by the fried product is important but also how the oil is distributed in the crust.

Bouchon, Aguilera and Pyle (2003) distinguished three oil fractions during the deep-fat frying process of potato cylinders: (a) structural oil – STO (absorbed during frying), (b) penetrated surface oil – PSO (suctioned during cooling), and (c) surface oil – SO. A small amount of oil penetrates during frying because most of the oil was picked up at the end of the process, suggesting that oil uptake and water removal are not synchronous phenomena. After cooling, oil was located either on the surface of the piece or suctioned into the porous crust microstructure. Pedreschi *et al.* (2008) found that the total oil content in potato chips was formed primarily by PSO (~89%), secondly by STO (~7%) and finally by SO (~4%). Most of the oil was absorbed by the potato chips in the first few minutes of frying. Oil absorption in potato chips is mainly

Table 9.1 Typical mean water and oil contents of some fried foods (source: Saguy and Dana 2003. Reproduced with permission of Elsevier)

Product	Mean water content (%)	Mean oil content (%)
Potato chips	2.5	34.6
Corn chips	1.0	33.4
Tortilla chips	1.8	26.2
Doughnuts (plain)	20.8	22.9
Onion rings	28.5	18.7
Chicken breast-breaded	45.7	18.1
Fish fillet-battered or breaded	53.6	12.9
French fries/par-fried	39.5/37.9	14.8/7.6

a surface phenomenon, which mostly takes place when the chips are removed from the fryer (during the cooling process). However, the percentages of distributions of each oil fraction could change according to pre-treatments (e.g. blanching, drying, etc.) or frying temperature (e.g. 120 or 180 °C) of the raw material. For instance, blanching of potato slices before frying could cause partial starch gelatinization originating a different microstructure with respect to the original one. These changes in microstructure could affect significantly not only the water migration pattern but also the uptake during frying (Pedreschi *et al.*, 2008). On the other hand, drying of raw materials before frying using microwave, hot-air treatment and baking have resulted in a significant reduction in oil content of different products (Moreira, Castell-Perez and Barrufet, 1999; Lamberg, Hallstrom and Olsson, 1990). Recently, much attention has been given to the use of hydrocolloid coatings such as methylcellulose, hydroxypropyl methylcellulose, long cellulose and corn zein to reduce oil uptake. The hydrocolloid mixture is usually added to the coating to create a barrier against oil absorption either before and/or during frying (Rimac-Brncic *et al.*, 2004; Williams and Mittal, 1999). Finally, vacuum frying may be an option for fried potatoes with low oil content and desired texture and flavour characteristics (Garayo and Moreira, 2002).

9.3 Properties of fried products

Frying is often selected as a method for creating unique flavours and texture in processed foods that improve their overall palatability. The high temperatures of the frying affect surface colour and mechanical characteristics of fried foods and heating of reducing sugars also influences a complex group of reactions, termed caramelization, leading to browning development, which defines the colour of the final product. Additionally, heat toxic compounds like acrylamide are formed during this process and their formation has been reported strictly linked to the Maillard reaction (Gökmen and Palazoglu, 2008; Pedreschi, Kaack and Granby, 2004).

Properties of raw or formulated food changes drastically during frying processes. A scheme of physical, chemical and structural changes occurring during frying of potatoes is shown in Figure 9.2. In the case of frozen products, water changes from the solid to the liquid phase throughout the sample and to the vapour (steam) phase in the outer, hot layers. After removal from the fryer, cooling steam condenses back to the liquid state. Heating up to 100 °C induces changes in starch and cells similar to those observed in cooking of potatoes. Starch granules undergo gelatinization at around 60–70 °C (e.g. they imbibe water and swell inside cells). In a similar temperature range (60–80 °C), the middle lamellae between cells disintegrates and cells get separated, giving the so-called mealy texture. Exposure to temperatures above 100°C causes starch granules and cells located in the forming crust to

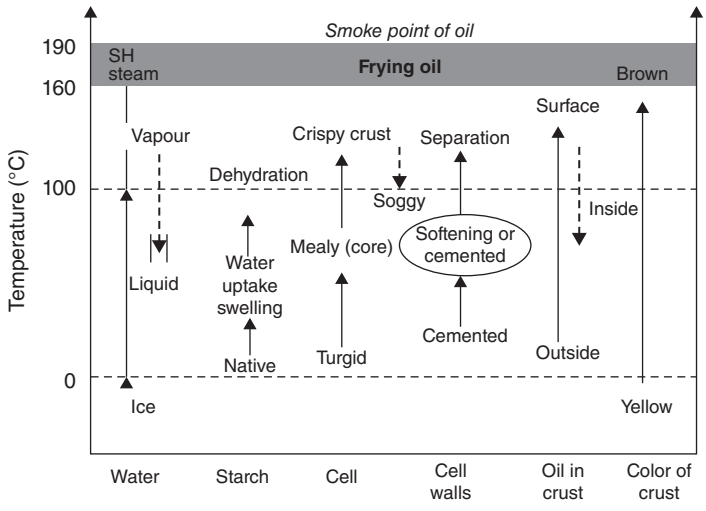

Figure 9.2 Physical, chemical and structural changes occurring during deep-frying of potatoes (source: Miranda and Agukera 2006. Reproduced with permission of Taylor & Francis Ltd.)

become dehydrated. Oil may then penetrate into crevices in the crust, but major impregnation occurs after frying and on cooling. The surface colour of products changes gradually with temperature to golden yellow and later to brown. Prolonged use of oil at high temperatures and in the presence of air leads to many reactions such as hydrolysis, oxidation, polymerization, etc. Some of the formed chemicals have been reported to pose health hazards. The term 'quality' of a fried product comprises a number of parameters of the frying material, either in a mid-state (at intermediate stages of the frying process) or after the completion of the frying. The properties of interest that determine the overall quality in food frying are moisture content, appearance/colour, textural properties, structure, oil content, toxic compound content (acrylamide, furan, etc.) and nutritional value.

9.3.1 Physical properties

Fat uptake is an important quality parameter of fried foods. Several factors affect oil uptake by fried products. The effect of frying temperature on oil uptake is unclear. Some authors state that frying at higher temperatures leads to a decrease in oil uptake, possibly due to the reduced frying time necessary, formation of a better developed crust, which may act as a barrier for oil absorption, or by a reduction in porosity of the crust (Baumann and Escher, 1995). Other authors have shown that the temperature effect on oil absorption is not significant (Pravisani and Calvelo, 1986). Surface roughness increases overall surface area, resulting in a higher oil uptake. Lately,

efforts have been made to reduce the fat content of deep-fried products and consequently lower their caloric density. The fat content of fried chips may be reduced using hot-air and superheated steam at the discharge end of the fryer, which removes nonabsorbed surface oil before it enters into the product. This procedure (low-fat stripping system) is claimed to reduce the oil content in chips by 25% (Kochhar, 1999).

The colour of potato chips is an extremely important criterion for the potato processing industry and is strictly related to consumer perception (Scanlon *et al.*, 1994). Consumers tend to associate colour with flavour, safety, storage time, nutrition, and level of satisfaction due to the fact that it correlates well with physical, chemical and sensorial evaluations of food quality. The final colour of potato chips is the result of the Maillard reaction, which depends on the content of reducing sugars and amino acids or proteins at the surface and the temperature and time of frying (Márquez and Añón, 1986). Browning becomes very rapid at temperatures higher than 150 °C and volatile flavour compounds are produced as secondary products. Treatments to lower the content of reducing sugars prior to deep-frying include exposing tubers to about 21 °C for 1–3 weeks, leaching sugars out from the surface with water or by blanching, exposing slices to a solution of the enzyme glucose-oxidase or by lactic fermentation with lactic bacteria (Halford *et al.*, 2012). The surface colour of potato chips is also highly dependent on the amount and distribution of the oil on the outer surface of the fried product.

Hindra and Baik (2006) have indicated that the changes in the colour parameters in fried foods followed first-order kinetics and the energy of activation (EA) for the total colour changes was 14–117 kJ/mol. The different magnitudes of the activation energy for colour changes were attributed to different product compositions, reaction mechanisms and estimation approaches. The texture of fried products is manifested when they are fractured, either experimentally or by mastication, and they may be analysed from the viewpoint of the fracture of materials. Materials contain flaws and cracks that act as stress concentrators, lowering their theoretical ultimate strength predicted from interatomic forces (Miranda and Aguilera, 2006). Crispness is a major textural property of fried foods derived from the low moisture content induced by frying either on the outer layers of thick pieces or throughout thin specimens. Crispness is a quality of brittle materials that rapidly fracture under stress at small strains and has been studied both by instrumental and sensory techniques (Roudaut *et al.*, 2002). The initial rising part of the force–deformation curve is a function of the stiffness of the sample. Stiffness depends on mechanical properties of the material and structural elements (e.g. air cells, microheterogeneities, geometrical parameters, etc.). The maximum force achieved before fracture is called hardness. In products that are heterogeneous at the microstructural level, as deformation increases

the phenomena repeat as a few smaller events of rise and sudden drop of the force, giving a jagged appearance to the force–deformation diagram. Small fractures are stopped by the presence of microheterogeneities, such as the presence of air cells, oil pockets and hard cells. This mechanical behaviour is known in textural terms as crispness.

Porosity and volume are also important physical properties in raw and processed vegetables because they are strongly linked to their mechanical properties and microstructure. For instance, a violent formation of bubbles is generated in potato tissue during frying, which determines the development of a microstructure with variable porosity in time that is affected by the operational parameters. A decrease in volume or shrinkage that takes place simultaneously during water vapour migration to the surrounding oil in potato during frying can affect transport properties such as thermal and mass diffusivities. Shrinkage and deformation depend on food geometry and method of frying.

Roughness is an important physical property that affects the processes of heat and mass transfer related to the food surfaces. Fractal analysis has been used to analyse the image texture of fried potatoes and to describe their surface microstructure quantitatively (Pedreschi *et al.*, 2012; Moreno, Brown and Bouchon, 2010; Pedreschi, Aguilera and Brown, 2000). Recently, some researchers have shown that the surface roughness of the sample to be fried is a key factor in oil absorption, but other food-related properties, such as the microstructure of the crust, may explain differences among product categories and should be examined in future studies (Moreno, Brown and Bouchon, 2010).

9.3.2 Structural features

Frying induces significant microstructural changes that can be understood by using different modern visualization techniques. For the case of potato chips, most revealing has been the video microscopy study of in situ frying of potato cells, which shows that the starch granules swell rapidly at a temperature of 60–70 °C, which is well below that of the frying. Steam bubbles leave the interior of the cells through pores in the cell walls (probably through plasmodesmata) and find their way to the product/oil interface through many intercellular passages. Swollen starch granules remained as a compact mass, pressing on the outer cell wall before becoming dehydrated at higher temperatures (Bouchon and Aguilera, 2001). Cells heated in oil to 180 °C remained largely intact and decreased marginally in surface area without any evidence of oil in their interior (Aguilera *et al.*, 2001). Confocal laser scanning microscopy (CLSM), a noninvasive technique that produces optical sections at increasing depths in the specimen, demonstrated that oil is placed as an egg-box arrangement surrounding intact potato cells (Figure 9.3).

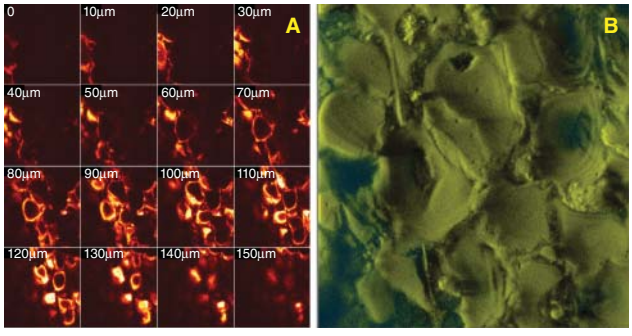

Figure 9.3 (A) Confocal images in the fluorescence mode of oil distribution in a potato chip fried in stained oil (170 °C, 3 min) observed at ×20. (B) Three-dimensional reconstruction from the serial sections in Figure 9.2 using Imaris software (source: Pedreschi *et al.*, 1999. Reproduced with permission of John Wiley & Sons, Ltd.). *See plate section for colour version*

9.3.3 Sensory attributes

The appearance and/or colour of the food surface is the first quality parameter evaluated by consumers even before it enters the mouth. Sensory evaluation of colour is frequently performed with application in quality control, product development and research. Sensory scientists reported that colour evaluations should be carefully carried out and pointed out the importance of the background colour in the viewing area, the kind of light source (and its intensity), the distance between the light source and the product, the panelists' viewing angle and the angle of light incidence on the sample to avoid specular reflection (Lawless and Heymann, 2010). Pedreschi *et al.* (2012) has showed how to classify potato chips according to their surface colour. The steps are (1) to design and to test a fast method for selecting proper judges based on colour ordering, (2) to establish quality categories of potato chips according to their surface colour and to apply these classification criteria in the colour evaluation of potato chips fried at different oil temperatures and after subjection to different pre-treatments and (3) to obtain models that will be able to describe the relation between the oil temperatures and pre-treatments employed with the frying times required to achieve the highest degree of quality.

9.3.4 Toxic compounds

Heating foods immersed in oil during frying can provide many advantages including improved taste, colour and texture and minimizing harmful germs. Further, flavour and aroma compounds are produced via the Maillard reaction, though some undesirable compounds may be produced; these are acrylamide, ethylcarbamate, furan, heterocyclic amines,

5-hydroxymethylfurfural (HMF), polycyclic aromatic hydrocarbons and nitrosoamines. Some compounds formed during frying of potatoes such as HMF, furan and a variety of Maillard reactants and lipid oxidation products may increase the risk of cancer for consumers. It is thus desirable to minimize the amount of these compounds (Pedreschi, 2012).

Acrylamide The presence of acrylamide in some fried and baked foods, most notably potato chips and French fries and baked cereal products, has been reported. Acrylamide has been classified by the International Agency for Research of Cancer (IARC) as 'probably carcinogenic for humans' and is recognized by the European Union Scientific Committee on Food (EUSCF) as a genotoxic carcinogen. However, no legal limits have yet been established for this contaminant in foods. Medeiros, Mestdagh and De Meulanaer (2012) summarized the studies on acrylamide levels, mechanisms of formation, assessment of acrylamide intake and health risk and possible mitigation strategies from farm to fork in fried potato products.

Acrylamide found in heat-processed food such as fried starchy foods is mainly formed from asparagine and a reducing sugar through Maillard reactions, which also generate compounds responsible for the brown colour and important flavour components of heated food (Mottram and Wedzicha, 2002; Stadler *et al.*, 2002). Thus, acrylamide is formed by the thermal reaction of natural components in starchy raw materials when moisture levels become low. However, acrylamide formation in starchy food is under kinetic control; this means that the amount that is formed depends on the rate of the reaction and the time for which it has been allowed to proceed. French fries and potato crisps exhibited relatively high values of acrylamide, such as 424 and 1739 ppb, respectively. Several factors, such as the initial concentration of the precursors, their ratio, heating temperature, time of processing, pH and water activity of the product influence the content of acrylamide in heat-processed foods (Yaylayan, Wnorowski and Pérez Locas, 2003).

Several technologies have been developed to reduce acrylamide concentration in thermally processed foods based on: (a) changing process parameters (e.g. time and temperature of cooking) which slows the Maillard reaction and (b) reducing acrylamide precursor levels in raw materials to be cooked at high temperatures (e.g. by using microorganisms, asparaginase, amino acids and saccharides, blanching, etc.).

Furan Furan is a cyclic ether with a boiling point of 31.4 °C, which has been identified as a potential human carcinogen that can be formed in a broad range of foods processed at high temperatures, such as coffee, baby foods, bread and snacks (International Agency for Research on Cancer, World Health Organization, 1995). Although it is still unclear what the risks are associated with the current intake levels of dietary furan, furan mitigation in foods may be considered a challenge in the prevention of human diseases, such as cancer

(Mariotti *et al.*, 2012). However, despite its high volatility, furan has also been found in low-moisture foods processed in open containers, such as potato chips and crackers. In contrast to acrylamide formation in foods, in which the Maillard reaction is the most important mechanism, furan formation in foods takes place by different pathways.

The reviews of some researchers described multiple pathways for the formation and occurrence of furan (Vranová and Ciesarová, 2009; Crews and Castle, 2007). The major pathways include thermal degradation or the Maillard reaction of reducing sugars, thermal degradation of some of the amino acids and thermal oxidation of ascorbic acid and polyunsaturated fatty acids. It is worthnoting that home and industrial cooked foods with high levels of carbohydrates (e.g. potato chips and French fries) are most likely to form furan, probably due to Maillard browning reactions of the food (Mariotti *et al.*, 2012). The furan content in fried products increases with an increase in the oil uptake levels.

Hydroxymethylfurfural 5-Hydroxymethylfurfural (HMF) can be regarded as one of the most important heat-induced contaminants occurring in starchy foods processed at high temperatures, such as potato chips and French fries. The amount of HMF detectable in foods is directly related to the heat load applied during processing of carbohydrate-rich products. HMF is a furanic compound that forms as an intermediate in the Maillard reaction and from direct dehydration of sugars under acidic conditions (caramelization) during thermal treatment applied to foods (Kroh, 1994; Ames, 1992). HMF occurs in high amounts in some selected foods and even if the mutagenic/carcinogenic activity is low, there might be a risk due to high exposure. This compound is formed by heating carbohydrate rich foods via the Maillard reaction. Higher temperatures, an acidic environment and low water activity favour the formation of HMF via the Maillard reaction. During the heat treatment, nonenzymatic browning reactions occur to develop the typical dark brown colour. The high temperature, low pH value and reducing water activity enhance the formation of HMF.

9.4 Machinery of frying

The most important aspects of industrial frying are the (a) type of frying oil, (b) nature of the food to be fried and its interaction with the frying oil, (c) frying equipment, (d) frying process, and (e) quality deterioration evaluation during frying. Commercial atmospheric fryers are extensively used in commercial kitchens and restaurants to cook snack foods, appetizers and specific entrees. These are generally made of thick stainless steel sheets to avoid corrosion over a longer period of use. Commercial fryers use different heating mechanisms such as infrared, convection, gas and electricity. Due to the high

cost of infrared and convection fryers, gas and electric fryers are the preferable options. While electric fryers have the advantage of mobility, gas fryers are generally more efficient and have a larger capacity than electric fryers. Gas fryers that use natural gas or propane can heat up to a high temperature quickly and have a faster temperature recovery time between frying cycles.

The tube style, open pot and flat bottom are the different types of common commercial atmospheric fryers. Most of the commercial fryers have several features such as a temperature control, a timer with an audible alarm, fry baskets of various shapes and sizes, automatic devices to lower and raise the basket out of the oil, castors, and oil filtration and ventilation systems. Fryer maintenance, regular inspections, cleaning and operator training are important for proper food quality, public health and to avoid potential hazards.

In this sense, continuous on-line fryers consists of a net belt conveying system with an elevating facility, automatic filtration system, oil temperature controlling system, oil addition and heating systems (Figure 9.4). This equipment is capable of frying different kinds of snacks either made from natural raw material or from dough pieces made from flour. Some advantages of the continuous fryers are a steady oil temperature, start-up automatic rising system for convenience in cleaning and maintenance, continuous operation at high efficiency, large output and reduced cost of production.

Fried foods continue to be popular in the restaurants since the fryer menu has extended to include various deep-fat fried snacks. Equipment manufacturers have responded by designing fryers that operate more efficiently, quickly, safely and conveniently. Fryers may be countertop units, free-standing floor units and in batteries of several fryers in one housing. Most of the fryers share a common basic design in which the kettle contains a sufficient amount of oil so

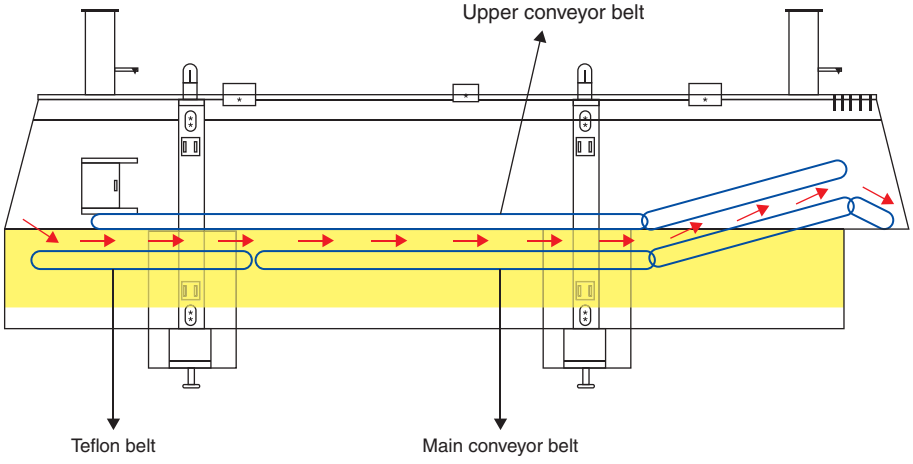

Figure 9.4 Setup of a continuous automatic fryer for snacks

that cooking is essentially supported by displacement of the oil rather than by the bottom of the vessel. The oil is typically heated by atmospheric or infrared gas burners underneath the kettles in 'fire tubes' that pass through the kettle walls. Electric fryers use heating elements immersed in the oil. Fryers range in capacity from 7 kg of oil for a small, and countertop fryer to over 90 kg of oil for the largest floor model fryers used for doughnut and chicken. Most fryers have a 'cold zone' at the bottom of the kettle where breadcrumbs and other particles settle. The cold zone is intended to have no convection current and a relatively low temperature, so that food crumbs will not carbonize and create the breakdown products that limit oil life. Finally, the type of energy and its method of delivering to the frying oil have some effect on oil degradation.

Aspects such as health, convenience and proper nutritional balance clearly demand process innovations. In this sense, vacuum frying is an efficient method of reducing the oil content in fried snacks, maintaining product nutritional quality and reducing oil deterioration. The unique aspect of vacuum frying is based on the fact that much lower temperatures (~120 to 130 °C) can be applied during frying. Thus, in vacuum frying operations, food is heated under reduced pressure (<60 torr or 0.0789 atm), causing a reduction in the boiling points of the oil and the moisture in the foods (Garayo and Moreira, 2002). These issues make this a suitable frying technology to produce fruits and vegetables with the necessary degree of dehydration without excessive darkening, scorching of the product or excessive loss of the flavours and natural colours. This kind of frying also has many other advantages over atmospheric frying, such as lower oil oxidation and the absence of frying vapour emissions.

Granda, Moreira and Tichy (2004) demonstrated that vacuum frying could produce fried starchy products with a much lower acrylamide content than their counterparts fried at atmospheric pressure. Industrially, a couple of Asian companies have developed a vacuum-fried system for the processing of fruit (apple, pineapple, grapes, banana, guava, mango, peach, etc.) and vegetables (sweet potato, potato, pumpkin, carrots, etc.) into chips as well as fried fishes and shellfishes (octopus and cattlefish). Due to the improvement in quality of the raw materials and blanching techniques, the use of vacuum fryers almost died out with the exception of one or two production companies who still insist in producing a nonblanched product. For certain applications, two-stage frying is applicable. In that case, the product is pre-fried in an atmospheric fryer and then subjected to vacuum frying until the final moisture content is reached. It is worth mentioning that several vacuum fryer units for the production of low-fat 'kettle-style' potato snacks are equipped with centrifuges for reducing the fat content of the samples after frying. The centrifuges are installed in a special vacuum dome attached to the vacuum fryer.

9.5 Stability of fried products

9.5.1 Water activity and sorption isotherms

Water plays an important role with respect to the properties of food systems. It influences the physical characteristics of fried products as well as their chemical stability. Moisture loss or gain from one region to another continues in order to reach thermodynamic equilibrium. The term activity of water (A_w) is used to indicate an intrinsic parameter of a food and equilibrium relative humidity (RH), a property of the surrounding atmosphere in equilibrium with the food system under consideration. The activity of water in a mixture can be expressed in terms of relative fugacity, defined as a measure of the tendency of a component to escape and is related to the chemical potential of the system (Enrione, Hill and Mitchell, 2007). Gal (1972) showed that in experimental terms there is small difference between the activity of water and the concept of equilibrium relative humidity (p_w/p_w^θ). Therefore A_w can be expressed as $A_w = (p_w/p_w^\theta)$, where p_w is the equilibrium water vapour pressure over the system and p_w^θ is the vapour pressure of pure water at the same temperature and pressure.

One of the most important outcomes of frying processing of foods is the reduction in its total moisture content and A_w (< 0.7), generating an unfavourable environment for spoiling microorganisms. Indeed, at frying temperatures of ~200 °C, water evaporates from the core of the material towards the surface, drying the food. Low values of A_w have been reported for pre-dried fried popped rice with values in the range of 0.3–0.4 (Phanitcharoen, Maliket and Siriwongwilaichat, 2010). Reis, Masson and Waszczynskyj (2008) reported values of A_w of about 0.5 for blanched potato sticks fried at 190 °C.

9.5.2 Sorption isotherms

The relationship between the total moisture content and the water activity, over a range of values at constant temperature, yields the sorption isotherm when expressed graphically. 'Equilibrium' is achieved when there is no water migration from/to the sample during storage. The adsorption isotherms can be classified into five general types, from which types I, II and III are more relevant to food systems. Type I is related to the Langmuir sorption behaviour, for example nonswelling porous solids, whilst type II is a combination of types I and III.

Most dried products that are being fried are expected to display their greatest stability at moisture contents comparable to the monolayer (Van den Berg, 1991). The initially hard and brittle material undergoes a glass transition, by the plasticizing effect of water, becoming weak, plastic or rubbery depending

on the polymer. Water molecules show a sharp increase in molecular mobility and diffusion, taking longer to reach 'equilibrium', as the rate of polymer swelling can become the limiting factor.

The sorption regions are useful in relating the rates of reaction for different fried food stability parameters to the water activity. Nevertheless, it must be considered that the term 'activity' is based on thermodynamic equilibrium, and most real foods do not reach this state between their various components nor with their environment. Indeed, the molecular mobility of the system increases as the moisture content increases, facilitating the encounter between reactants to interact in the system (e.g. reducing sugar and amino groups in nonenzymatic browning).

9.5.3 Modelling sorption isotherms

The sorption process at low partial vapour pressures can be described as 'a localized physical adsorption on an initially rigid adsorbent'. However, the hydrophilic nature of most food materials and the plasticizing effect of water at higher relative humidity promote a gradual increase in the system's mobility, which can lead to the solubilization of the polymer (Enrione, Hill and Mitchell, 2007).

Many mathematical relations have been proposed to represent sorption isotherms in food matrices. Two parameter equations have been proposed to describe the sorption process by Hasley, Smith, Henderson and Brunauer–Emmett–Teller. To improve the fitting throughout a wider range of water activities, three and four parameter equations have also been developed. From this group, the equations of, for example, the Brunauer–Emmett–Teller (BET) model and the Guggenheim–Anderson–de Boer (GAB) model have been applied due to their theoretical derivation, which provides quantitative information associated to the adsorption phenomenon. Other equations with four fitting parameters have been proposed by Peleg (1993) and Crapiste and Rotstain (1982). For the former, the mathematical structure and number of constants were chosen to show that the sorption sigmoid shape at low RHs is mainly determined by a process of a decreasing rate with respect to RH, similar to the Langmuir sorption kinetics (Enrione, 2005). The equation of Crapiste and Rotstain considers the chemical potential of water at each phase of the food system to predict the amount of water at equilibrium. Hickey *et al.* (2006) described a sorption isotherm for potato chips after relative humidity equilibration (40 to 95%) at 25 °C, which was modelled using the BET equation (Figure 9.5a). Kawas and Moreira (2001) used the Crapiste and Rotstain approach to predict the desorption isotherm at three different temperatures (25, 48.8 and 68.8 °C) of tortilla chips fried in vegetable oil (Figure 9.5b). In the case of fried chips the amount of total oil content was also measured. The amount of water at equilibrium in the dough and fried chips was lowered with an increase in

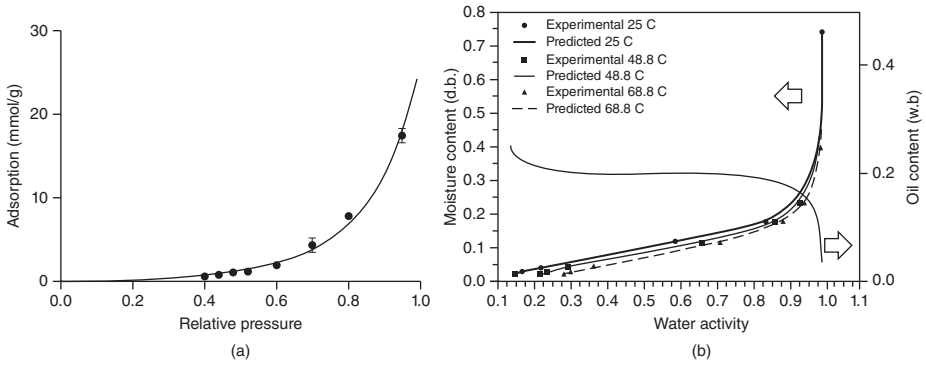

Figure 9.5 (a) Absorption isotherm obtained for potato chips (source: Hickey *et al.*, 2006. Reproduced with permission of Elsevier), and (b) desorption isotherm obtained for tortilla fried chips with the amount of total oil (secondary axis) (source: Adapted from Kawas and Moreira 2001. Reproduced with permission of Elsevier)

temperature. This behaviour can be attributed to the exothermal nature of the sorption process; hence an increase in temperature would not favour water–matrix interactions.

9.5.4 Glass transition temperature

Food polymers and the behaviour of their mixtures are mainly responsible for the structure–properties relationship in foods. The two basic features of food are that its biopolymers, such as proteins and polysaccharides, are its main construction materials, and water is the main medium, solvent and plasticizer (Tolstoguzov, 2008). Foods matrices can form amorphous or partially amorphous structures when the solvent (water) is removed at different rates. This process can occur during dehydration by temperature in processes that involve heating or cooling (freezing). The supercooled amorphous fraction exists in the nonthermodynamic equilibrium state and exhibit time-dependent changes associated to viscous flow towards the equilibrium state (Roos, 2009). This nonequilibrium state produces variations in the molecular structuring and molecular mobility.

The glass transition temperature or T_g, which occurs over a temperature range, has been defined as a single temperature at which, on rapid cooling, an amorphous material becomes extremely viscous ($\sim 10^{12}$ Pa s) (Roos, 1995). Mechanically the material behaves as a solid but maintains its amorphous structure as a liquid, therefore containing excess in free energy. At temperatures below T_g, the molecular mobility is restricted to vibrations and short-range rotational motions (Slade and Levine, 1995). The relationship between T_g and moisture content of amorphous biopolymers (food major constituents) is an important one, as it can determine the kinetic of deteriorative processes during isothermal storage. Indeed, the

plasticizing effect of water can reduce T_g, altering the molecular mobility of the matrix. If T_g is lower than the ambient temperature the amorphous fraction present in the food will behave as a rubber, allowing sufficient mobility for deteriorative processes such as reactant-driven reactions and structure recrystallization (Roos, 1995). Kingcam, Devahastin and Chiewchan (2008) studied the degree of starch retrogradation of frozen potato chips processed by blanching (ratio of potato to water of 0.015 g/g at 90 ± 2 °C for 5 min) and various freezing–thawing cycles. They observed a significant increase in retrogradation (%) from 20.05 to 29.12% after blanching, cooling and five freezing–thawing cycles.

If the product moisture content is reduced, T_g of the product will increase at higher temperatures than the ambient temperature, becoming rigid and brittle. Despite the increase in viscosity of the system in the glassy state, structural changes would still occur at the macro scale, such as densification, a phenomenon known as physical ageing (Hutchinson, 1995).

A number of expressions have been proposed to estimate the glass transition temperature of amorphous mixtures based in their compositions. However, the different equations can all be represented as minor variations of the same mathematical form (Pinal, 2008), where the T_g of the food mixture can be related to the glass transition temperature and the weight fraction of each component in association with fitting constants that account for the change in heat capacity between the rubbery and glassy states (ten-Brinke, Karasz and Ellis, 1983; Couchman and Karasz, 1978; Gordon and Taylor, 1952).

T_g in fried products is a very important parameter as it can determine the extent of processing, product physical and sensory properties and stability. Figure 9.6 illustrates an example of T_g values of tortilla chips with the total and partial (internal) oil content as a function of the moisture content (Kawas and Moreira, 2001). The continuous line represents the numerical fitting of

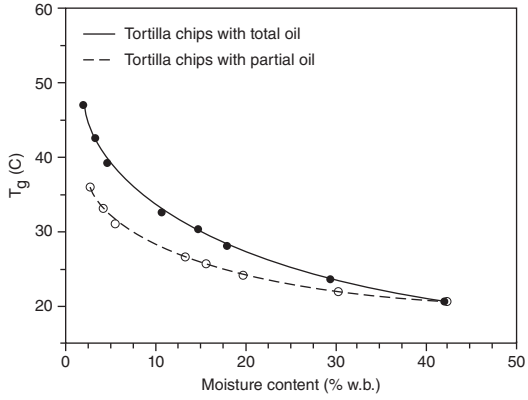

Figure 9.6 Glass transition temperature of tortilla chips (total and partial oil contents) fried for 60 s at 190 °C (source: Kawas and Moreira 2001. Reproduced with permission of Elsevier)

the Gordon–Taylor equation (Gordon and Taylor, 1952) to the experimental data. Interestingly, differences were detected between the T_g of the total and partial oil content samples. This was attributed to the hydrophobic nature of the oil and possibly the reduction of gelatinization of starch by formation of amylose lipid complexes, which reduces the overall plasticizing effect of water in the matrix.

9.5.5 Glass transition and water diffusion in foods

During absorption or desorption of water, foods undergo transitions that depends on temperature and molecular mobility of the polymer (Enrione, Hill and Mitchell, 2007). Water diffusion in glassy and rubbery states can be mathematically described by the classic Fickian approach. Indeed, in the glassy state, molecular mobility is restricted due to the increase in viscosity of the system (Roos, 1995) and the relaxation kinetics of the polymer chains. Therefore, the diffusion process becomes concentration dependent. In the case of the rubbery state, the molecular relaxation becomes instantaneous compared to the kinetics of water diffusion, becoming also concentration dependent following the Fick diffusion model (Enrione, Hill and Mitchell, 2007). Complexity arises near the glass transition zone where the relaxation time of the polymer structure becomes relevant. Fick's law is no longer applicable without the inclusion of a time–stress term accounting for viscoelastic relaxation of the polymer, which can oppose or enhance the fluid movement in the fluid in the polymer matrix (Takhar, 2008). Indeed, Pedreschi and Moyano (2010) modelled the diffusion of water in potato chips during frying by variable effective diffusion coefficients (D_{eff}) (~0.002 to 0.020 mm^2/s) after 4.5 minutes of frying at 150 °C, acknowledging the complexity of the frying process.

The understanding of water diffusion mechanisms is very important for the correct assessment of the oil uptake kinetics in food matrices during frying as it has been related to the removal of water by evaporation during this unit operation. Indeed, Ziaiifar *et al.* (2008) have mentioned that water loss is an explanatory variable for structural transformations and oil uptake because water escape is at the origin of very diverse material phenomena.

9.6 Conclusions

Deep-fat frying is a rather complex process comprising simultaneous heat and mass transfer with chemical reactions and textural changes taking place. The food sample in contact with hot oil heats up and water is lost and the oil content increases; the reaction between reducing sugars and amino acids leads to browning and changes of texture, with softening at the beginning of frying and hardening of the external layers of food with a longer frying time.

Additionally, deep-fat frying processes can take place at atmospheric pressure (conventional frying) or at pressures below atmospheric (vacuum frying). This complex unit operation also involves significant microstructural changes; in fact, most of the desirable characteristics of fried foods are derived from the formation of a composite structure: a dry, porous, crispy and oily outer layer or crust, and a moist cooked interior or core.

Frying is often selected as a method for developing unique flavours and improving texture in processed foods that enhance their overall palatability. The high temperatures of frying typically leads to the appreciated surface colour and mechanical characteristics of fried foods; heating of reducing sugars affects a complex group of reactions, termed caramelization, leading to browning development, which defines the colour of the final product. Additionally, toxic compounds (like acrylamide) are formed during this process and its formation has been reported as being strictly linked to the Maillard reaction. Moreover, some heat-induced toxicants that can be formed during frying are ethylcarbamate, furan, heterocyclic amines, 5-hydroxymethylfurfural (HMF), polycyclic aromatic hydrocarbons and nitrosoamines. First moisture content and then oil content play major roles in the chemical and microbiological stability of fried products. Moisture isotherms and the glass transition temperature (T_g) are very important tools to understand the behaviour of fried structures during storage. For instance, crispy structures of fried snacks have T_g values much higher than the temperature of the environment.

Acknowledgement

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References

Aguilera, J.M., Cadoche, L., Lopez, C. and Gutierrez, G. (2001) Microstructural changes of potato cells and starch granules heated in oil. *Food Research International*, **34**, 939–947.

Ames, J.M. (1992) The Maillard reaction. In: *Biochemisrty of Food Proteins* (Hudson, B.J.F., ed.), vol. **4**, Chapter 4. London: Elsevier Applied Science, London, England, pp. 99–153.

Baik, O.D. and Mittal, G.S. (2003) Kinetics of tofu color changes during deep-fat frying. *Lebensmittel-Wissenschaft und Technologie*, **36**, 43–48.

Baumann, B. and Escher, E. (1995) Mass and heat transfer during deep fat frying of potato slices. I. Rate of drying and oil uptake. *Lebensmittel-Wissenschaft und Technologie*, **28**, 395–403.

Bouchon, P. and Aguilera J.M. (2001) Microstructural analysis of frying potatoes. *International Journal of Food Science and Technology*, **36**, 1–8.

Bouchon, P., Aguilera, J.M. and Pyle, D.L. (2003) Structure oil-absorption relationships during deep-fat frying. *Journal of Food Science*, **68**, 2711–2716.

Bouchon, P., Hollins, P., Pearson, M., Pyle, D.L. and Tobin, M.J. (2001) Oil distribution in fried potatoes monitored by infrared microspectroscopy. *Journal of Food Science*, **66**, 918–923.

Couchman, P.R. and Karasz, F.E. (1978) A classical thermodynamic discussion of the effect of composition on glass-transition temperatures. *Macromolecules*, **11**, 117–119.

Crapiste, G.H. and Rotstein, E. (1982) Prediction of sorptional equilibrium data for starch-containing foodstuffs. *Journal of Food Science*, **47**, 1501–1507.

Crews, C. and Castle, L. (2007) A review of the occurrence, formation and analysis of furan in heat processed foods. *Trends in Food Science and Technology*, **18**, 365–372.

Danna, D. and Saguy, I.S. (2006) Mechanism of oil uptake during deep-fat frying and the surfactant effect – theory and myth. *Advances in Colloids and Interface Science*, **128–130**, 267–272.

Enrione, J. (2005) Mechanical stability of intermediate moisture starch–glycerol systems. PhD Thesis, Division of Food Sciences, The University of Nottingham, England.

Enrione, J., Hill, S. and Mitchell, J. (2007) Sorption behaviour of mixtures of glycerol and starch. *Journal of Agricultural and Food Chemistry*, **55**, 2956–2963.

Farkas, B.E., Singh, R.P. and Rumsey, T.R. (1996) Modeling heat and mass transfer in immersion frying. 1. Model development. *Journal of Food Engineering*, **29**, 211–226.

Gal, S. (1972) Expression of concentration of water vapour in water vapour sorption measurements. *Helvetica Chimica Acta*, **55**, 1752–1757.

Garayo, J. and Moreira, R. (2002) Vacuum frying of potato chips. *Journal of Food Engineering*, **55**, 181–191.

Gökmen, V. and Palazoglu, T.K. (2008) Acrylamide formation in foods during thermal processing with a focus on frying. *Food and Bioprocess Technology*, **1**, 35–42.

Gordon, M. and Taylor, J.S. (1952) Ideal copolymers and the second order transitions of synthetic rubbers. I. Non-crystalline copolymers. *Journal of Applied Chemistry*, **2**, 493–500.

Granda, C., Moreira, R. and Tichy, S.E. (2004) Reduction of acrylamide formation in potato chips by low-temperature vacuum frying. *Journal of Food Science*, **68**, 405–411.

Halford, N.G., Curtis, T.Y., Muttucumaru, N., Postles, J., Elmore, E. and Mottram, D.S. (2012) The acrylamide problem: a plant and agronomic science issue. *Journal of Experimental Botany*, **63**, 2841–2851.

Hickey, H., MacMillan, B., Newling, B., Ramesh, M., Van Eijck, P. and Balcom, B. (2006) Magnetic resonance relaxation measurements to determine oil and water content in fried foods. *Food Research International*, **39**, 612–618.

Hindra, F. and Baik, O.D. (2006) Kinetics of quality changes during food frying. *Critical Reviews in Food Science and Nutrition*, **46**, 239–258.

Hutchinson, J.M. (1995) Physical aging of polymers. *Progress in Polymer Science*, **20**, 703–760.

International Agency for Research on Cancer (IARC), World Health Organization (WHO) (1995) *Dry-Cleaning, Some Chlorinated Solvents and Other Industrial Chemicals*, vol. 63, IARC Monographs on the Evaluation of the Carcinogenic Risks to Humans. IARC, WHO, Lyon, France, p. 89.

Kawas, M.L. and Moreira, R.G. (2001) Chracterization of product quality attributes of tortilla chips during the frying process. *Journal of Food Engineering*, **47**, 97–107.

Kingcam, R., Devahastin, S. and Chiewchan, N. (2008) Effect of starch retrogradation on texture of potato chips produced by low-pressure superheated steam drying. *Journal of Food Engineering*, **89**, 72–79.

Kochhar, S.P. (1999) Safety and reliability during frying operations – Effects of detrimental components and fryer design features on frying of food. In: *Principles, Changes, New Approaches* (Varela, G., Bender, A.E. and Morton, I.D., eds). Ellis Horwood Ltd, Chichester, England.

Kroh, L.W. (1994) Caramelisation in food and beverages. *Food Chemistry*, **51**, 373–379.

Lamberg, I., Hallstrom, B. and Olsson, H. (1990) Fat uptake in a potato drying/frying process. *Lebesmittel-Wissenschaft und Technologie*, **23**, 295–300.

Lawless, H. and Heymann, H. (2010) *Sensory Evaluation of Food*, 2nd edn. Springer, New York, USA, 596 pp.

Mariotti, M., Granby, K., Fromberg, A., Risum, J., Agosín, E. and Pedreschi, F. (2012) Furan occurrence in starchy food model systems processed at high temperatures: effect of ascorbic acid and heating conditions. *Journal of Agricultural and Food Chemistry*, **60**, 10162–10169.

Márquez, G. and Añón, M.C. (1986) Influence of reducing sugars and amino acids in the colour development of fried potatoes. *Journal of Food Science*, **51**, 157–160.

Medeiros, R., Mestdagh, F. and De Meulanaer, B. (2012) Acrylamide formation in fried potato products – Present and future, a critical review on mitigation strategies. *Food Chemistry*, **133**, 1138–1154.

Mellema, M. (2003) Mechanism and reduction of fat uptake in deep-fat fried foods. *Trends in Food Science and Technology*, **14**, 364–373.

Miranda, M. and Aguilera, J.M. (2006) Structure and texture properties of fried potato products. *Food Reviews International*, **22**, 173–201.

Moreira, R.G., Castell-Perez, M.E. and Barrufet, M.A. (1999) *Deep-Fat Frying: Fundamentals and Applications*. Aspen Publishers, Maryland, USA.

Moreno, M.C., Brown, C. and Bouchon, P. (2010) Effect of food surface roughness on oil uptake by deep-fat fried products. *Journal of Food Engineering*, **101**, 179–186.

Mottram, D.S. and Wedzicha, B.L. (2002) Acrylamide is formed in the Maillard reaction. *Nature*, **419**, 448–449.

Moyano, P. and Pedreschi, F. (2006) Kinetics of oil uptake during frying of potato slices: effect of pre-treatments. *Lebensmittel-Wissenschaft und Technologie*, **39**, 285–291.

Ngadi, M., Li, Y. and Oluka, S. (2007) Quality changes in chicken nuggets fried in oils with different degrees of hydrogenation. *Lebensmittel-Wissenschaft und Technologie*, **40**, 1784–1791.

Pedreschi, F. (2012) Frying of potatoes: physical, chemical, and microstructural changes. *Drying Technology*, **30**, 707–725.

Pedreschi, F., Aguilera, J.M. and Arbildua, J.J. (1999) CLSM study of oil location in fried potato slices. *Microscopy and Analysis*, **37**, 21–22.

Pedreschi, F., Aguilera, J.M. and Brown, C.A. (2000) Characterization of food surfaces using scale-sensitive fractal analysis. *Journal of Food Process Engineering*, **23**, 127–143.

Pedreschi, F., Aguilera, J.M. and Pyle, L. (2001) Textural characterization and kinetics of potato strips during frying. *Journal of Food Science*, **66**, 314–318.

Pedreschi, F., Kaack, K. and Granby, K. (2004) Reduction of acrylamide formation in potato slices during frying. *Lebensmittel-Wissenschaft und Technologie*, **37**, 679–685.

Pedreschi, F. and Moyano, P. (2005) Oil uptake and texture development in fried potato slices. *Journal of Food Engineering*, **70**, 557–563.

Pedreschi, F. and Moyano, P. (2010) Effect of water content on physical properties of potato chips. In: *Water Properties in Food, Health, Pharmaceutical and Biological Systems: ISOPOW 10* (Reid, D.S., Sajjaanantakul, T., Lillford, P.J. and Charoenrein, S., eds), 1st edn. Blackwell Publishing, Iowa, USA.

Pedreschi, F., Moyano, P., Hernández, P. and Figueroa, C. (2005) Modeling water loss during frying of potato slices. *International Journal of Food Properties*, **8**, 289–299.

Pedreschi, F., Cocio, E., Moyano, P. and Troncoso, E. (2008) Oil distribution in potato slices during frying. *Journal of Food Engineering*, **87**, 200–212.

Pedreschi, F., Bunger, A., Skurtys, O., Allen, P. and Rojas, X. (2012) Grading of potato chips according to their sensory quality determined by color. *Food Bioprocess and Technology*, **5**, 2401–2408.

Peleg, M. (1993) Assessment of a semi-empirical four-parameter general model for sigmoid moisture sorption isotherms. *Journal of Food Process Engineering*, **16**, 21–37.

Phanitcharoen, S., Maliket, A. and Siriwongwilaichat, P. (2010) Effect of drying and frying time on textural and sensory characteristics of popped rice. *Asian Journal of Food and Agro-Industry*, **3**, 368–372.

Pinal, R. (2008) Entropy of mixing and the glass transition of amorphous mixtures. *Entropy*, **10**, 207–223.

Pravisani, M.F. and Calvelo, A. (1986) A minimum cooking time for potato strip frying. *Journal of Food Science*, **51**, 614–617.

Raoult-Wack, A.L., Vitrac, O., Trystram, G. and Lucas, T. (2000) Water mediated phenomena in some food processes. In: *Water Science for Food, Health, Agriculture and Environment: ISOPOW 8* (Berkn, Z., Leslie, R.B., Lillford, P.J. and S. Mizrahi, S., eds). Technomic, Lancaster, pp.241–254.

Reis, F.R., Masson, M.L. and Waszczynskyj, N. (2008) Influence of blanching pretreatment on color, oil uptake and water activity of potato sticks, and its optimization. *Journal of Food Process Engineering*, **31**, 833–852.

Rimac-Brncic, S., Lelas, V., Rade, V. and Simundic, B. (2004) Decreasing of oil absorption in potato strips during deep fat frying. *Journal of Food Engineering*, **64**, 237–241.

Roos, I. (1995) *Phase Transition in Foods*, 1st edn. Academic Press Inc., San Diego, California, USA.

Roos, I. (2009) Mapping the different states of food components using state diagrams. In: *Modern Biopolymer Science, Bridging the Divide between Fundamental Treatise and Industrial Application* (Kasapis, S., Norton, I.T. and Ubbink, J., eds). Academic Press, London, UK.

Roudaut, G., Dacremont, C., Valles Pamies, B., Colas, B. and Lemeste, M. (2002) Crispness: a critical review on sensory and material science approach. *Trends in Food Science and Technology*, **13**, 217–227.

Saguy, I. and Danna, D. (2003) Integrated approach to deep fat frying: engineering, nutrition, health and consumer aspects. *Journal of Food Engineering*, **56**, 143–152.

Scanlon, M.G., Roller, R., Mazza, G. and Pritchard, M.K. (1994) Computerized video image analysis to quantify color of potato chips. *American Potato Journal*, **71**, 717–733.

Slade, L. and Levine, H. (1995) Glass transitions and water-food structure interactions. *Advances in Food and Nutrition Research*, **38**, 103–269.

Sosa-Morales, M.E., Orzuna-Espíritu, R. and Vélez-Ruiz, J.F. (2006) Mass, thermal and quality aspects of deep-fat frying of pork meat. *Journal of Food Engineering*, **77**, 731–738.

Stadler, R.H., Blank, I., Varga, N., Robert, F., Hau, J., Guy, P.A., Robert, M.C. and Riediker, S. (2002) Acrylamide from Maillard reaction products. *Nature*, **419**, 449–450.

Takhar, P.S. (2008) Role of glass-transition on fluid transport in porous food materials. *International Journal of Food Engineering*, **4**, DOI: 10.2202/1556-3758.

ten-Brinke, G., Karasz, F.E. and Ellis, T.S. (1983) Depression of glass transition temperature of polymer networks by diluents. *Macromolecules*, **16**, 244–249.

Tolstoguzov, V. (2008) Food polymers in food material science. In: *Food Materials Science, Principle and Applications* (Aguilera, J.M. and Lillford, P.J., eds). Springer, New York, USA.

Van den Berg, C. (1991) Food-water relations: progress and integration, comments and thoughts. In: *Water Relations in Foods* (Slade, L. and Levine, H., eds). Plenum Press, New York, USA.

Vélez-Ruiz, J.F. and Sosa-Morales, M.E. (2003) Evaluation of physical properties of dough of donuts during deep-fat frying at different temperatures. *International Journal of Food Properties*, **6**, 341–353.

Vranová, J. and Ciesarová, Z. (2009) Furan in food – a review. *Czech Journal of Food Science*, **27**, 1–10.

Williams, R. and Mittal, G.S. (1999) Low-fat fried foods with edible coatings: modeling and simulation. *Journal of Food Science*, **64**, 317–322.

Yaylayan, V.A., Wnorowski, A. and Pérez Locas, C. (2003) Why asparagine needs carbohydrates to generate acrylamide. *Journal of Agricultural and Food Chemistry*, **51**, 1753–1757.

Ziaiifar, A., Courtois, F. and Trystram, G. (2010) Porosity development and its effect on oil uptake during frying process. *Journal of Food Process Engineering*, **33**, 191–212.

Ziaiifar, A., Achir, N., Courtois, F., Trezzani, I. and Trystram, G. (2008) Review of mechanisms, conditions, and factors involved in the oil uptake phenomenon during the deep-fat frying process. *International Journal of Food Science Technology*, **43**, 1410–1423.

10

Roasting and Toasting Operations in Food: Process Engineering and Applications

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10.1 Introduction

Roasting or toasting is a typical dry heating food processing operation having different objectives to meet certain specific requirements. The main aims here are to cook (without using water or oil) or gelatinize, expand/pop/puff the food materials, inactivate antinutritional factors and present the food products in a more amenable, palatable and appealing form. There is no universally accepted definition for roasting while its demarcation with toasting is unclear and appears to be related to products. For example, the commonly used term roasting is related to grain, coffee and nut (Fellows, 2009) while toasting is associated with cereal flakes (Fast, 1990). However, a differentiation can be drawn between roasting and toasting by considering the time–temperature combination. In general, roasting is a process where the temperature is lower but the time is higher (may be more than several minutes) while toasting is conducted at a much higher temperature for a short duration (say less than one minute). Until the late nineteenth century, dry heat roasting in an oven was called baking. However, roasting is one of the oldest forms of cooking meat in an open fire.

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Toasting is an important processing method (Table 10.1) to produce ready-to-eat low-fat snacks. Toasting is thus an alternative to deep-fat frying. Usually air is the most suitable medium for toasting and the method implies a high temperature for a short time whereas roasting means a slow heating process for longer duration. However, both of these unit operations are a process of moisture removal through a simultaneous heat and moisture transfer phenomenon in foods. A group of flavour compounds called carbonyls was developed during roasting (Toledo and Brody, 1999). They usually participate in reactions that result in roasted flavours through the formation of large complex molecules that typically have a yellow or brown colour and the characteristic roasted flavour. Flavour development through the Maillard reaction depends on the concentration of reducing sugars and amino acids in the food, moisture content, temperature and time of treatment. The reaction appears to be favoured by high temperature and low moisture content. In high moisture containing samples, the moisture evaporation from the outer surface is adequately replaced by diffusing water from the interior, resulting in a high moisture level at the surface. Hence, the Maillard reaction and flavour development occur only when a dry surface crust is formed. On the other hand, roasting at high temperatures rapidly forms the dry surface crust, resulting in an intense Maillard reaction and flavour development compared to low temperature roasting. The flavour of coffee and cocoa, roasted nut and beef may be attributed to the reactions involving these carbonyls. Although fat itself has no flavour, its presence in roasted foods intensifies the surface temperature from radiant heating in the oven, and the hydrophobic nature of fat favours the Maillard reaction and flavour development more compared to the same product without fat (Toledo and Brody, 1999).

10.2 Applications of the process in specific foods

A number of food ingredients and products are subjected to dry roasting or toasting without using oil and water. The list includes meat, fish, tea, coffee, grains, cocoa, etc.

10.2.1 Coffee

Raw or green coffee is roasted and finally powdered for using as a beverage. The desirable typical aroma or flavour is developed during roasting. Pressure develops in the coffee beans due to heat and that pressure holds the initial breakdown products together until the proper stage of roasting is reached; at this latter stage, they react with each other and produce the characteristic flavour. Rocha *et al.* (2003) have identified over 850 flavour-related substances in the volatile fraction of the roasted coffee. Obviously, roasted coffee flavour largely depends on the manner and extent of roasting. Freshly roasted beans

Table 10.1 Some roasting and toasting processes

Type of processing	Raw material	Specific material	Machinery employed	References
Toasting	Grain	Corn flake	Fluidized air toaster	Sumithra and Bhattacharya (2008)
Roasting	Meat	Beef muscle	Domestic electrical oven	Goni and Salvadori (2010)
Roasting	Meat	Turkey breast	Steam convection oven	Bialobrzewski *et al.* (2009)
Roasting	Bean	Coffee beans	Fluidized bed hot-air roaster	Schenker *et al.* (2002)
Roasting	Nut	Hazelnut	Laboratory batch air oven	Demir *et al.* (2003)
Roasting	Nut	Peanut	Pilot scale cross-flow hot-air roaster	Davidson, Brown and Landman (1999)
Roasting	Nut	Almond	Commercial fluidized bed hot-air roaster	Zhang *et al.* (2011)
Roasting	Nut	Cashew nut	Hot cast iron pans	Fagbemi (2008)
Roasting	Bean	Coffee	Lab scale coffee roaster	Hecimovic *et al.* (2011)
Roasting	Seed	Lotus seed	Microwave roasting	Bhat and Sridhar (2011)
Roasting	Seed	Cranberry seed	Thermostat controlled hot plate	Aremu *et al.* (2010)
Roasting	Nut	Almond	IR roasting, sequential infrared and hot air (SIRHA) roasting and traditional hot-air (HA) roasting	Yang *et al.* (2010)
Roasting	Meat	Beef steaks	Oster toaster oven and kitchen oven	Shen *et al.* (2011)
Roasting	Seed	Bambara groundnut	Hot fine sand in a pan	Ijarotimi and Esho (2009)
Roasting	Meat	Ducks	Large oven using smokeless hard wood fuel	Chen, Song and Ma (2009)
Roasting	Cereal	Buckwheat	Oven	Zhang *et al.* (2010)
Roasting	Cereal	Buckwheat	Microwave oven	Zhang *et al.* (2010)
Roasting	Nut	Chestnut	Heated electrical plate	Nazzaro *et al.* (2011)
Roasting	Root	Sweet potato roots	Spherical grill with heated charcoal	Kidmose *et al.* (2007)

Table 10.1 (*continued*)

Type of processing	Raw material	Specific material	Machinery employed	References
Roasting	Cereals, legumes, nuts	Finger millet, amaranth, pigeon pea, field bean, groundnut, pumpkin, sunflower, butternut	Open iron pan container using charcoal burner	Kunyanga *et al.* (2011)
Roasting	Bean	Green coffee bean	Fluidized bed hot-air roaster	Wang, Fu and Lim (2011)
Roasting	Bean	Locust bean	Oven	Akinoso and Raji (2011)
Roasting	Millet	Little millet	Electrically heated bowl roaster	Pradeep and Guha (2011)
Roasting	Bean	Red kidney beans, pinto beans, black-eyed peas and soybeans	Microwave oven	Boateng *et al.* (2008)
Roasting	Fish	Marine fishes in Nigeria	Roasted over hot charcoal	Oluwaniyi, Dosumu and Awolola (2010)
Roasting	Poultry meat	Chicken breast, duck breast	Oven roasting	Liao *et al.* (2010)
Roasting	Meat	Iberian pig muscle	Oven roasting	Broncano *et al.* (2009)
Roasting	Meat	Iberian pig muscle	Microwave	Broncano *et al.* (2009)
Toasting	Nut	Almond	Household toaster oven	Kong and Singh (2009)
Roasting	Bean	Velvet bean	Microwave	Bhat, Sridhar and Bhushan (2007)
Roasting	Nut	Hazelnut	Microwave	Uysal, Sumnu and Sahin (2009)

give the best flavour and aroma. To obtain quality coffee, roasted beans should be ground freshly prior to consumption rather than keeping the ground coffee longer.

10.2.2 Cocoa

Fermented and dried cocoa beans are roasted for development of the characteristic flavour of cocoa. Roasting helps in two ways in cocoa processing. It causes changes in the chemical structure of polyphenols, producing less astringent compounds, and helps in the development of the characteristic cocoa flavour. Roasting also makes the outer shell brittle and helps in removing them (Manay and Shadaksharaswamy, 2008).

10.2.3 Popping of cereals

Roasting leads to popping of food grains when subjected to high temperature for a short time, utilizing the entire grain including the outermost husk, bran and germ. The sudden application of high temperature expands the moistened grains to several folds to yield a low-density product (such as 100 kg/m^3). The pericarp thickness of the grain is an important indicator of popping quality and is largely influenced by the genetic factor. The popping quality including volume expansion is influenced by the moisture content of the grain and the time and temperature of roasting. Roasting here introduces the desirable porous and crunchy texture and develops an agreeable caramelized/aromatic flavour. The high temperature of roasting produces bigger and uniform air cells and leads to an adequate expansion (Manay and Shadaksharaswamy, 2008). Examples of pop cereals are popped rice (Figure 10.1, left), popcorn, etc.

Figure 10.1 Photograph of popped rice (left) and puffed rice (right)

10.2.4 Puffing of cereals

Roasting or toasting involves high-temperature treatment for a short period, resulting in a sudden expansion of water vapour in the interstices of the granule of a cereal grain to make it expand or puff (Figure 10.1, right). Unlike popping, puffing can be conducted on grain or grain fractions, and is not necessarily a one-step process phenomenon like popping. Puffing is not necessarily a varietal-dependent process. The grain or grain fraction has to be moistened, conditioned and roasted in such a way that it expands during the high-temperature short-time roasting process. The high temperature of roasting even induces cell rupture and cell separation.

Popping or puffing of grains or grain fractions is conducted by oven or gun puffing, or by roasting with sand, salt or hot air. In both roasting and toasting operations, the mode of heat transfer is by conduction (using sand, salt or even a hot metal surface), convection (using hot air), radiation (using infrared/microwave) or a combination of these types of heat transfer. To avoid nonuniform expansion, further processing may be required to make the product attractive (Manay and Shadaksharaswamy, 2008). Temperature has a tremendous effect on the microstructure and texture of puffed grain. Low temperature gives nonuniform and small air cells. On the other hand, high-temperature puffing shows bigger and more uniform air cells. The highly porous microstructure of popped rice, puffed rice and popcorn shows the air cells in Figure 10.2.

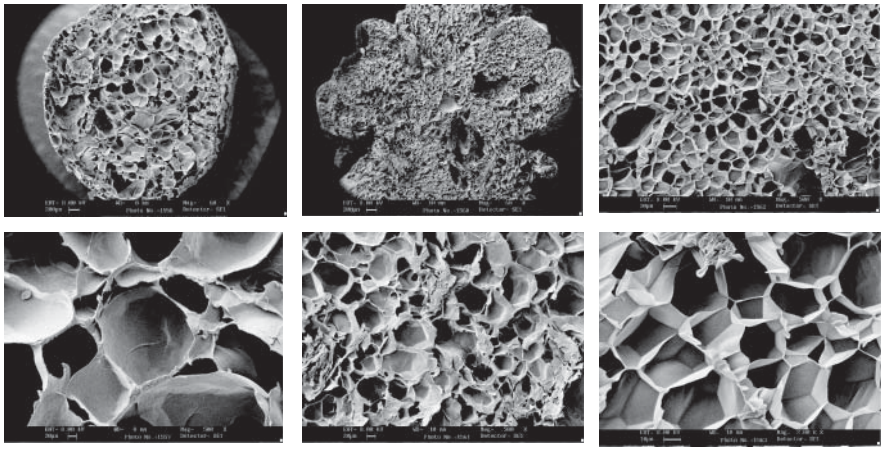

Figure 10.2 Top row shows the low magnification views of the cross-sections of puffed rice (left), popped rice (centre) and popcorn (right). Bottom photomicrographs are the corresponding high magnification views of the same samples showing cellular structures

10.2.5 Toasting of breakfast cereals and snacks

Toasting is an integrated processing step in the manufacture of several breakfast cereals and snacks. Flattened or flaked cereals having 10 to 15% moisture content is toasted (blistered) at a high temperature (250–300 °C) for a short time (about 30 s) in a rotary toaster oven. Corn, rice, sorghum, wheat or millets are cleaned, conditioned and cooked under pressure with added flavourings followed by flaking and toasting to make ready-to-eat breakfast cereals. The final toasting step is also called blistering as this step helps the flaked or flat cereals to puff or create blisters on the outer surface. This process makes the product crispy and reduces the density. The temperature and time of toasting are the crucial factors for the quality of the finished product and consumer acceptance (Sumithra and Bhattacharya, 2008). Overtoasting or undertoasting operation adversely affects the quality of breakfast cereals. The toasting operation also helps in developing the desired colour, flavour and texture. Snack foods are often subjected to the roasting/toasting operation as a finishing step to bring some specific changes like attaining a low moisture content and inducing crispness. Examples include cereal/pulse-based ready-to-eat snacks and other low-fat snacks such as pillow-shaped puffed rice crispy (Figure 10.3), where the process of frying is replaced by the roasting/toasting process.

A decrease in the bulk density of a product due to puffing offers a new organization of the outer layers and high porosity of the matrix. A rapid hydration of the puffed materials occurs due to capillary liquid absorption. Raw breakfast flake loses its uniform good appearance due to toasting. Several tiny bulged portions are created in products offering an additional crisp and

Figure 10.3 Pillow-shaped low-fat puffed rice snack

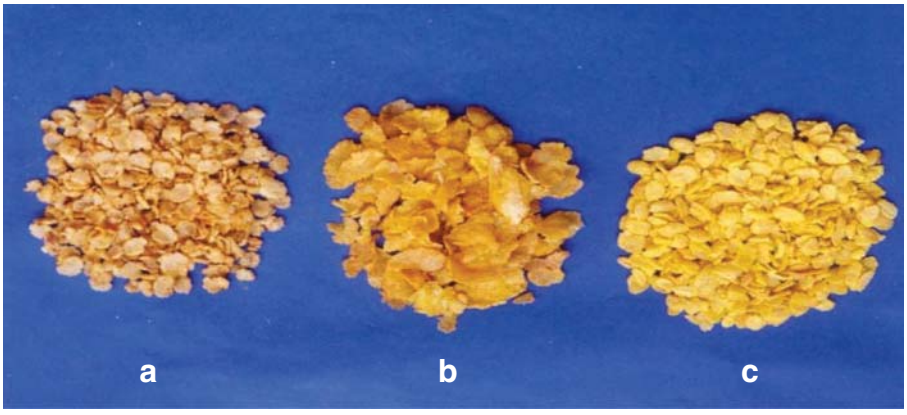

Figure 10.4 Breakfast cereals from (a) sorghum, (b) corn and (c) rice. *See plate section for colour version*

soft texture. Though corn flake is an internationally accepted breakfast cereal, other cereal flakes are also possible (Figure 10.4).

The microstructure of the untoasted corn flake (Figure 10.5a) appears to be a dense material having only a few small pores (Sumithra and Bhattacharya, 2008). On toasting, the thickness of the flakes increases and a porous structure results (Figure 10.5b). These large vacuoles are possibly formed by the fusion of a number of pores and are usually elongated in shape so that the toasted flakes become soft and crispy. Heating with hot air causes rapid evaporation of moisture, resulting in the creation of a number of small pores. As heating continues, these pores grow rapidly and fuse with other pores to create large vacuoles. These pores and vacuoles cannot collapse any further; they decide the final texture of the toasted flakes as the evaporation of moisture in flakes is fast and the final moisture content is low. The toasted flakes may show a

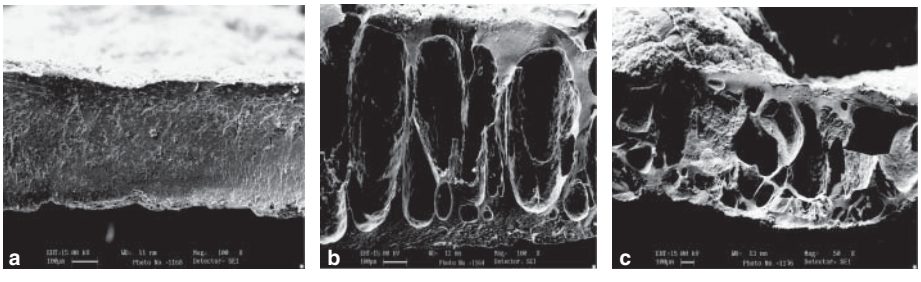

Figure 10.5 Photomicrograph of (a) an untoasted raw corn flake, (b) a toasted corn flake at an appropriate moisture, temperature and time and (c) a toasted corn flake with a collapsed structure because of high-temperature toasting (source: Sumithra and Bhattacharya 2008. Reproduced with permission of Elsevier)

few extra bulged portions, indicating localized high expansion (Figure 10.5c). However, this particular zone appears to possess a soft texture with a crispy mouth feel. The thickness of the flakes and pore volume decide the retention of crispness when breakfast cereals are soaked in milk or juice for serving.

10.2.6 Roasting of pulse/legume

Roasting of pulse and/or legume has been practised in Asia, the Middle East and South America for many years. The whole pulse/legume is exposed directly (or after addition/sprinkling with a small quantity of water) to dry heat in a roasting pan with pre-heated sand or pre-heated edible salt. Depending on the type of legume, the pan temperature is maintained at 200 to 250 °C and the sample is heated for 2–3 min. Sand or salt is then separated by sieving after roasting. Roasted chickpea is a popular traditional snack in several countries. During roasting, water inside the chickpea causes an increase in the vapour pressure, leading to expansion, and gives the roasted chickpeas a porous structure and chalky appearance. Along with gelatinization of starch in roasted pea, denaturation of protein also occurs in addition to inactivation of enzyme and antinutritional factors, and enhances the product shelf-life. Roasted pea flour can be effectively used as a flavour carrier or an improver, and can be incorporated into several food formulations (Sumati, Shalini and Rajagopal, 2006).

10.2.7 Roasting of spice

Roasting can intensify the flavour of spices along with inactivation of contaminating organisms. Roasted spices are liked by the majority of people because of the development of attractive flavours. Roasting of whole spice as well as roasting of powdered spice are practised.

10.2.8 Toasting of bread

Bread becomes brown when toasted by exposure to radiant heat. Toasting also helps stale bread to become firmer and more palatable. Bread toasters are common domestic small ovens for toasting bread by placing it into the narrow slots on the top of the toaster and then using a lever.

10.2.9 Roasting of meat

Meat roasting can be achieved by using dry heat either by an open flame, in an oven or by using other heat sources. Slower cooking of meat in a bulk quantity occurs in an oven due to diffused heat. Since hot air circulates around the meat, cooking is done on all sides uniformly. Roasting of meat can be

done using low-temperature cooking, high-temperature cooking and a combination of both depending on the food and the tastes of the people. Slow roasting between 95 and 160 °C is the usual temperature range for cooking of meat and offers less moisture loss and a more tender final product. For high-temperature roasting (i.e. above 200 °C), the water inside the muscle is lost at a high rate and there will be browning on the outer surface of the product; a variety of flavour develops while the centre may be undercooked. In the combination method, high heat just at either the beginning or the end of the cooking process is used. A golden brown texture and crust formation occurs and maintains more of the moisture than simply cooking at a high temperature. The temperature of roasting of meat is a critical factor; a lower roasting temperature for a longer period of time is considered better than a higher temperature for short periods of time. To minimize the loss of moisture from meat, butter, lard or oil can be applied on the exterior surface. The multiobjective optimization of beef roasting has been performed by considering the minimization of both the cooking time and weight loss, and a mathematical model has been developed (Goni and Salvadori, 2012).

Meat pieces are placed uncovered on a rack in a shallow pan. The roasting pan is then placed at the centre of the oven, maintained at 165–170 °C. A thermometer records the temperature at the centre of the thickest part of the meat and roasting is continued until the desired internal temperature is attained. This ensures the adequate browning of meat with good flavour and appearance. Several changes occur due to roasting meat in addition to the inactivation of microorganisms. Colour changes of the pigment in meat during roasting indicate the extent of doneness. Well-done meat is more denatured and browner. During roasting, some volatile breakdown products are produced from proteins and amino acids of the meat. Lipid components are also broken down into several volatile compounds. All these volatile compounds are responsible for the overall flavour and taste of the finished product.

10.2.10 Roasting of nut

Nuts are eaten as raw, roasted, steamed or fried, although roasted nuts are preferred. Nuts are oven roasted or hot-air toasted or salt roasted. To prepare groundnut flour, mild roasting is required to separate the skin and to remove the germ. Finally, after extraction of oil from the kernels, it is ground into flour. However, to add consumer appeal and to be fit for product formulation, shelled nuts undergo processes including blanching, roasting or grinding. Since nuts are susceptible to fungi, contamination results in multiplication of microorganisms and in the production of mycotoxins. Hence, during roasting of nuts, one should ensure that every nut reaches some minimum temperature; at the same time, it gives the product a crisp texture and appealing

colour. As a result of roasting, the moisture content in nuts decreases from an initial value of 6–8% down to 2–3%. The high fat content in nuts limits their shelf-life. Shelling of cashew nuts is usually done by roasting at 200–250 °C for 120–180 s. The roasting process adds flavour and taste to the nuts. However, underroasting adversely affects the quality as well as the recovery of the nut kernel. After roasting, the edible portion of the nuts is carefully separated to avoid breakage. Finally, the product is flavoured and salted, followed by grading based on their size. Nuts are roasted to a moderate degree and the skin and the germ are removed during the preparation of peanut butter. Properly roasted kernel is then ground with the addition of salt, sugar and oil.

Roasting is one of the simplest and oldest processes to remove the outer seed coat or hull from certain tree legume seeds like the tamarind seed. Direct sand roasting is a commonly used age-old industrial technique. Tamarind kernel powder (TKP) is mostly used for sizing purposes in the textile and jute industries and the seed coat is removed completely. The seeds are roasted with sand at a considerably high temperature (100–250 °C) for 2 to 3 min for the testa to become loosened from the kernel so that it can be easily removed by rubbing in a decorticator followed by aspiration-separation. The time and temperature of roasting are the most important process variables that affect the subsequent steps like de-hulling and grinding of kernels. The colour of the kernel powder varies widely between brown to white depending on the time and temperature of roasting. Examination of the seed microstructure gives an insight into the morphological changes occurring during different heat and mass transfer processes and helps in selecting the appropriate conditions for processing. The raw seed (Figure 10.6a) shows the presence of three different layers – the outer most seed coat followed by an aleurone layer and the cotyledon or endosperm. In the raw seed, these layers are firmly attached to one another. The approximate thickness of the seed coat of a matured seed is between 150 and 250 μm. Figure 10.6b and 10.6c shows the cross-sections of the sand-roasted (225 °C for 2 min) tamarind seed, where loosening of the seed coat occurs due to roasting, which facilitates the next de-hulling process.

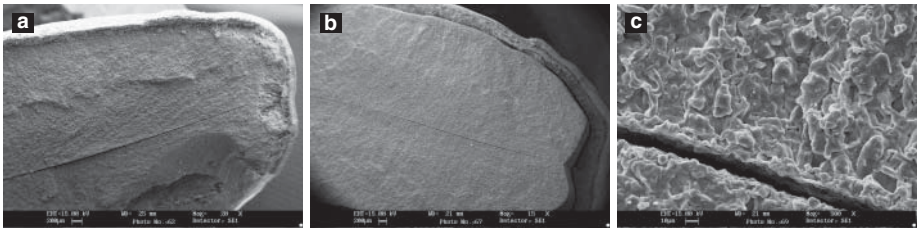

Figure 10.6 Scanning electron photomicrographs of the cross-sections of (a) a raw tamarind seed, (b) a sand-roasted seed, and (c) magnified view of sample shown in (b)

10.3 Process modelling

The modelling of the roasting/toasting process linking product attributes is a difficult job. Several approaches have been attempted. These are kinetic studies (Davidson, Brown and Landman, 1999; Bhattacharya and Prakash, 1997), an empirical approach such as response surface methodology (RSM) (Sumithra and Bhattacharya, 2008) and the heat/mass transfer approach (Scher, Fazio and Hsieh, 1993).

The effect of the food roasting/toasting process on the product is a result of heat and mass transfer, which can be characterized by the rate equations (Bhattacharya and Prakash, 1997). The mathematical representations are based on the law of conservation of mass and energy. Hence, the concentration of the reactants and effect of temperature on the reaction rates are important for material and energy balances. One easily applied statistical technique for estimating the process parameters is called a step-wise multiple regression (SMR). The input to the SMR consists of the independent variable $(X_1, X_2, \ldots, X_n)$ and the dependent variable (Y) in the following form:

$$Y = c_0 + c_1X_1 + c_2X_2 + \ldots + c_nX_n + E \tag{10.1}$$

where c_0, c_1, c_2, $\cdots$, c_n are the coefficients determined by the SMR and E is the error. Once the mathematical model is established, it should be validated for its applicability. The fit of the model can be judged by the correlation coefficient and the magnitude of the residual or variance of the dependent variables. However, second-order polynomials can also be used instead of the first-order regression model shown in Equation (10.1). Interpretation of the model graphically leads to better understanding of the process and optimization of the processing conditions.

The effect of toasting variables such as moisture content of the corn flakes, temperature and time of toasting on quality attributes has been investigated (Sumithra and Bhattacharya, 2008). The response functions studied are the thickness of flake, bulk density, puncture force and sensory attributes (appearance, texture and overall acceptability). These response functions are correlated well with the independent variables by second-order polynomials. It is observed that temperature and time of toasting have more effect over the moisture content of the corn flakes. The optimum condition for the best puffing and highest overall acceptability is obtained at a moisture content of 11.2%, a temperature between 250 and 257 °C and time between 39 and 45 s. On toasting, the flakes do not only puff by its thickness but also show a porous microstructure such that the toasted flakes possess a good crisp texture (Sumithra and Bhattacharya, 2008).

Process analysis tools including simulation and mathematical modelling may be useful to optimize the design of process equipment and control parameters of the simultaneous heat and mass transfer-based operations.

The major mechanisms for mass transfer primarily include diffusion, while the enthalpy transfer mechanisms include conduction and convection. The important factors considered for dry roasting operation are a roasting medium such as air, inert gas, indirect or direct heating mode, determination of batch versus continuous operation, operating temperature, energy input source, humidity and velocity range. The characteristics of the food material such as flowability, abrasiveness, size, shape, density, melting point/flash point, upstream/downstream processing requirements, production capacity and target attributes are also important in the decision-making process for selection of the type of roasting system.

The most used dry cooking performance by a convection system (Scher *et al.*, 1993) is defined by the maximum drying rate (N_w):

$$N_w = \frac{h_{\text{conv}}\, \dot{a} V\, (T_w - T_g)}{Q_v} \tag{10.2}$$

where $h_{\text{conv}}\, \dot{a}$ is the convective volumetric heat transfer coefficient, T_g is the bulk gas temperature, T_w is the wetted product surface temperature, V is the product volume and Q_v is the rate of heat transfer per unit mass of moisture. Equation (10.2) is valid only for an unhindered drying rate where food structure does not inhibit the drying rate. The convective heat transfer coefficient is determined from existing semi-empirical relations for impingement surfaces (Scher, Fazio and Hsieh, 1993):

$$N_{\text{Nu}} = a\, {N_{\text{Re}}}^{b} \tag{10.3}$$

where N_{Nu} is the dimensionless Nusselt number, N_{Re} is the dimensionless Reynolds number and a and b are constants.

Thermal efficiency (N_{T}) for convection-based dry cooking systems is defined as

$$N_{\text{T}} = \frac{(\text{T}_{\text{supply}} - T_{\text{return}})}{(T_{\text{supply}} - T_{\text{return}} + (1 - R_{\text{e}})\, (T_{\text{return}} - T_{\text{amba}}))} \tag{10.4}$$

where T_{supply} is the supply air temperature, T_{return} is the return air temperature, T_{amba} is the ambient air temperature and R_{e} is the air recirculation ratio. An analysis of the mass transfer rate versus thermal efficiency is necessary to govern the air recirculation ratio. The three commonly used performance indices for heat and mass transfer unit operations are (a) specific heat consumption, calculated from the ratio of the amount of heat supplied to the mass of water evaporated, (b) specific power consumption, the ratio of energy consumed to the food material throughput, and (c) specific volume, determined as the ratio of the dry cooking chamber volume to the food material throughput. To

control heat and mass transfer resistances, the Biot numbers (Scher, Fazio and Hsieh, 1993) are defined by

$$N_{\mathrm{BiH}} = \frac{h_{\mathrm{conv}} I}{k} \quad \text{and} \quad N_{\mathrm{BiM}} = \frac{h_{\mathrm{conv\ mass}} I}{D} \tag{10.5}$$

where I is a characteristic transfer dimension, k is the thermal conductivity of food and D is the moisture diffusivity. The terms N_{BiH} and N_{BiM} are the Biot number for heat transfer and the Biot number for mass transfer, respectively, and h_{conv} and $h_{\mathrm{conv\ mass}}$ are the convective heat transfer coefficient and the convective mass transfer coefficient, respectively.

The convective mass transfer coefficient is determined as follows (Scher, Fazio and Hsieh, 1993):

$$h_{\mathrm{conv\ mass}} = \frac{h_{\mathrm{conv}}}{\left(C_{p_a}\ \rho_a \left(\frac{N_{\mathrm{Sch}}}{N_{\mathrm{Pr}}} \right)^{2/3} \right)} \tag{10.6}$$

where C_{pa} and ρ_a are the specific heat and mass density, respectively. The terms N_{Sch} is Schmidt number and N_{Pr} is Prandtl number.

In the roasting or toasting unit operation, the dynamic and steady-state model simulations can provide the relationships between the equipment design parameters, process design parameters and food attributes along with the design and analysis of the processing operations.

The enthalpy balance for the individual compartment physical system can be formulated by summing all the stream enthalpy contributions at the combustion of the flue gases entry point. The different process parameters that impact the inlet supply air temperature of the food sample include the flame temperature, ambient/make-up air temperature and humidity, recirculation air flow ratio, inlet/supply air humidity, food granules bed outlet air temperature and humidity, inlet supply air flow rate and combustion flue gas flow rate.

A fuzzy control system can represent the food processing operations including roasting and toasting by providing the knowledge that is derived from numerical simulations, empirical observations or by their combinations. For continuous cross flow roasting of a peanut, a simulation study on the effect of heating rate, peanut size, ratio of peanut mass loading to air mass flow ($G_{\mathrm{p}}/G_{\mathrm{a}}$) and roasting air temperature has been conducted (Davidson, Brown and Landman, 1999). Fuzzy rules describe the effects of variations on peanut size, $G_{\mathrm{p}}/G_{\mathrm{a}}$ and air temperature on the time to reach a minimum roasting temperature. The fuzzy logic controller includes feedforward and feedback components to ensure compatibility of the two elements. Feedforward control is based on the process model for the kinetics of peanut heating and colour changes during roasting. A threshold temperature has to set which is defined experimentally as the temperature above which the

rate of browning due to Maillard reactions becomes appreciable. The change in lightness value (L^*) with the time for peanut temperatures above the threshold temperature is described by a zero-order kinetic model (Davidson, Brown and Landman, 1999).

The variability in temperature during the roasting of hazelnuts, two models of the unsteady-state heat transfer based on an analytical solution for solid sphere geometry and on a numerical solution for hollow sphere geometry, has been proposed by Demir *et al.* (2003). Variations in the hazelnut temperature are predicted using theoretical formulas and the Monte Carlo method. For optimizing the roasting process with respect to uniformity in product quality and safety, the prediction of variability in the nut roasting temperature is important. Further research is required to determine the effect of roasting temperature on the quality of hazelnuts.

For multiobjective optimization of meat roasting, a validated mathematical model has been exploited considering the minimization of both cooking time and weight loss (Goni and Salvadori, 2010). An increase in oven temperature from 120 to 225 °C significantly reduces the cooking time. A positive relation exists of cooking weight loss with cooking time and/or with increased oven temperature. Oven energy consumption also increases when the oven temperature is higher.

10.4 Machinery and methods

Roasting or toasting of coffee beans or nuts or grains are conducted by batch and continuous roasters. Foods are heated directly by contacting with flame or indirectly by employing infrared/microwave systems and rotary drum roasters. In drum-type roasters, beans or nuts move from the overhead feeding hopper into a cylindrical drum that turns to keep the beans in constant motion for uniform heating. The drum can be heated by circulating heated air or with radiant heat from the drum walls, the exterior of which can be heated by contact with hot air, gas flame or steam. Frequently, the drum-type roaster is replaced with tunnel or fluidized bed-type ovens in which the beans or nuts pass on moving belts or are vibrated beneath an infrared bulb radiating heat source under controlled temperature. The commonly employed roaster/toaster is discussed in the subsequent section.

10.4.1 Puffing gun

Puffing guns are frequently used for snacks and breakfast cereals to obtain a crisp texture and toasted flavour; it is one of the oldest methods of puffing cereals. Flattened cereals or grains are placed in a puffing gun where the materials are heated under pressure to convert the moisture within the food materials into steam. The gun is then opened suddenly and the steam under

pressure within the food materials expands explosively and puffs the kernels (Fast, 1990). The method of gun puffing is also used to half-puff the cereal dough pieces or extruded moist pellets. These half-puffed materials are again hot-air toasted to develop the toast-flavoured ready-to-eat snacks.

10.4.2 Batch roaster

The basic system of a batch coffee roaster (Figure 10.7) consists of (a) a hopper for feeding the material, (b) a roasting chamber or drum through which hot air passes and the material gets mixed, (c) a discharge unit that unloads the roasted material at the end of roasting and (d) a driving motor. The roasting chamber is generally a horizontal drum, which at a desired angle can be rotated for good mixing, uniform roasting and discharge.

A batch roaster with recirculation of air is also possible by employing a furnace that supplies the heated air by burning a fuel gas or fuel oil. A blower delivers air to the burner in the furnace and suitable valves control the rate at which fuel and air flow is needed. A cyclone may be installed for collecting the chaffs and husks that are generated during roasting. Another blower recirculates most of the hot gas for energy efficiency.

10.4.3 Continuous roaster/toaster

Continuous systems (inclined horizontal systems and spiral tube systems) are employed for large-scale manufacturing of roasted grains, nuts, oil seeds,

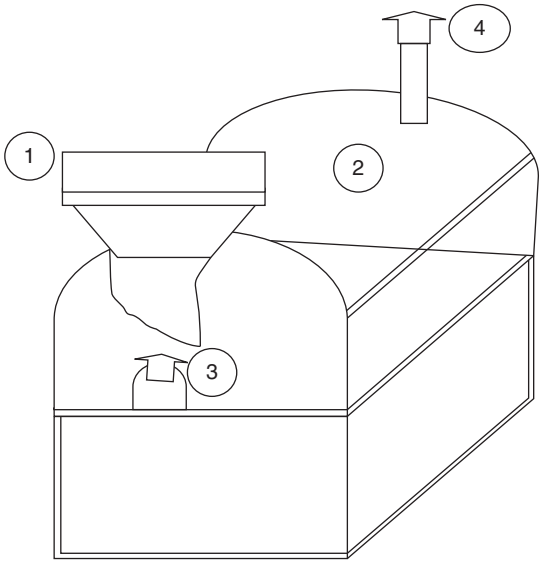

Figure 10.7 Schematic view of a batch roaster: 1, feeding hopper, 2, rotating drum, 3, product exit gate and 4, discharged gas

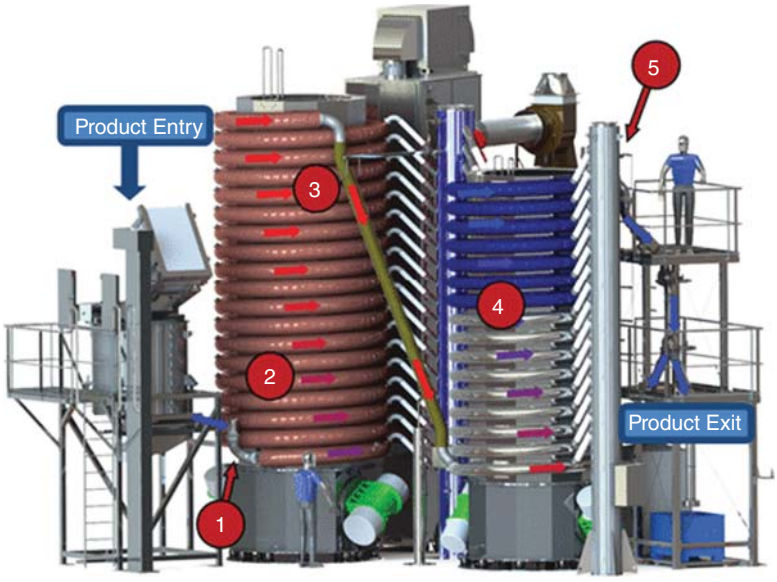

Figure 10.8 Continuous spiral roaster: 1, feeding system, 2, vibratory hot tubes to transport materials, 3, roasted materials progressing tubes, 4, cooling tubes and 5, product exit (source: REVTECH, France. Reproduced with permission of REVTECH Process Systems). *See plate section for colour version*

coffee and cocoa seeds. There are three phases for a continuous spiral roaster (Figure 10.8), which consist of (a) feeding of the raw material at a constant flow rate, (b) materials are heated in the spiral tubes directly for a desired residential time and temperature where the material is gradually roasted and transported to the next cooling chamber by vibration and (c) roasted material is allowed to cool in the third phase of second spiral tubes where injection of cool and dry air reduces the temperature of the material as well as stabilizes the product to be suitable for packaging. A wide range of products can thus be roasted or toasted, such as almonds, cashew nuts, pistachios, hazelnuts, sunflower seeds, pumpkin seeds, sesame seeds, wheat, barley, tea and spices. A temperature as high as 300 °C and production capacity of 5000 kg/h can be achieved.

10.4.4 Fluidized bed roaster

Heated air is blown upwards through the food particles with just enough force to suspend the particles in a gentle boiling motion. Heated air is introduced through a porous plate that supports the bed of the food materials (Figure 10.9). The moist air exists at the top. The time for roasting/toasting mainly depends on the depth of material, moisture content and temperature of roasting.

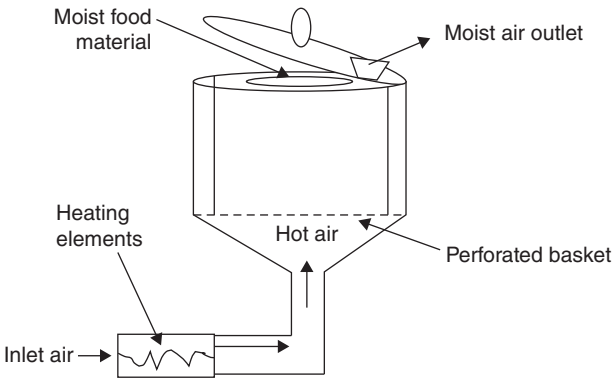

Figure 10.9 Batch-type fluidized bed roaster

In the case of a continuous-type fluidized bed roaster, the material moves on a vibratory perforated metallic belt through which hot air passes at a moderate speed to help fluidization. Adjustment of a suitable inclination also aids the movement of the material during heat treatment.

10.4.5 Microwave roasting

Microwave roasting or toasting is the quickest method of heat treatment of food materials. Microwaves with a frequency of 950 or 2450 MHz are generated by a magnetron. The limit of penetration of microwaves to food is only a few centimetres. An advantage of microwave roasting is that the time of roasting is ten times faster than a conventional cooking method, saving considerable time and electrical energy (Potter and Hotchkiss, 1995). A limitation of microwave roasting is that the crust formation or browning of outer surface does not occur in the roasted food and consequently the flavour development is also poor. The specific applications of microwave roasting/toasting are for developing roasted cereals (Zhang *et al.*, 2010), legumes/beans (Boateng *et al.*, 2008), meat (Broncano *et al.*, 2009), etc.

Eichner (1999) describes a method and machinery for roasting coffee beans that provide the rapid heat transfer with precise control of roasting time–temperature profiles by monitoring process conditions, such as inlet and outlet gas enthalpies. A patented device has been reported for impinging steam on to the food item while moving on a conveyor along a toasting pathway. The device has been claimed to be suitable for steaming and toasting sandwich bun heels and crowns (Ewald and Simmons, 2011). An edge toasting shield (blocking IR waves) has been designed for toasting English muffins (Veltrop and D'van, 2011). The edge shield is a baffle formed of orthogonal or substantially orthogonal metal strips while the other one is

a cylindrical tube, so that the IR waves hit the food product edges at angles of incidence less than 80 degrees for obtaining a better-quality product.

10.5 Changes during roasting/toasting

The dry cooking food unit operation involves high-temperature dehydration systems and several physical and chemical changes occur during the high-temperature treatment. Roasting manifests a profound change in the moisture content, appearance, taste, aroma and texture, and inactivates enzymes and microorganisms, enhancing the shelf-life of the product and also changing the nutritional value of the product. This unit operation is associated with chemical reactions such as the Maillard reaction, sugar caramelization, protein denaturation/degradation, starch gelatinization and pyrolysis of the various organic constituents (Fellows, 2009). The other associated changes are appearance/colour (Fellows, 2009), texture (Wattanachant, Benjakul and Ledward, 2005), flavour development (Adrian, 1982) and inactivation of antinutritional factors (Ari *et al.*, 2012), which finally affect consumer acceptability. During cooking of meat, the structure of collagen breaks down to a soft gelatin, becomes soft and separation of muscle fibre occurs, allowing juice to come out of the meat to yield the dried meat. Over-roasting can cause muscle fibres to shrink and become tough because of dried-out tissues (Wattanachant, Benjakul and Ledward, 2005).

Several chemical reactions take place between sugars, proteins and minerals during roasting and the breakdown of hydroxyl amino acids, and degradation of pigments occurs, yielding several types of volatile compounds such as sulfur compounds, pyrazines, pyridines, pyrroles, oxazoles, aldehydes, ketones, phenols, etc., along with carbon dioxide. The beta-damascenone, 2-furfurylthiol (or furfurylmercaptan), 3-mercapto-3-methylbutyl formate, 2,5-dimethyl-4-hydroxy-3[2H]-furanone and guaiacol are some of the important aroma compounds responsible for the characteristic aroma in roasted coffee (McGorrin, 2006).

Hazelnut samples have been roasted at 135 °C for different times, and the results show that linoleic acid, essential amino acids and total sugar contents decrease significantly after 20 min of roasting. It is concluded that roasting for 15 min at 135 °C is suitable for maintaining the desired nutritional value of hazelnuts (Kirbaslar and Erkmen, 2003).

Roasting significantly influences the nutrient composition of the coconut meal as well as the physicochemical properties of the extracted oil. The colour of oil from roasted seeds is brown while the oil extracted from raw coconuts is colourless. The aroma and taste of both oils from roasted and raw coconut are pleasant. Saponification value for roasted coconut seed oil increases by

5 times compared to raw sample, while markedly increasing the flash point. Increasing trend is observed for the iodine and peroxide values, but low free fatty acid occurs in the roasted coconut seed oil (Amoo, 2004).

An improved breakfast cereal biscuit method has been described by Adrian, Ann and Marcus (2000) comprising the grain and a waxy grain fraction. The grain has been hydrated and cooked sequentially, rolled into flakes and either agglomerated followed by toasting into a desired biscuit shape or toasted followed by agglomeration into the targeted shape. These improved breakfast cereal biscuits have been claimed to possess more nutritional value, a better shelf-life, a tender crisp texture and better flavour, and require less energy to manufacture than standard flaked wheat breakfast biscuits.

Toasting helps in desolventization of spent soybean oilseed meal by using a conventional desolventizer toaster (DT) (Kemper, 2000). The DT has been designed to introduce enough heat to evaporate the solvent from the soybean meal. This can be achieved by using a direct or indirect steam heating process and the introduction of more countercurrent trays, increasing of steam flow, etc. Designing a DT with an optimal configuration for solvent stripping and meal quality can be achieved by controlling the pressure drop in countercurrent trays.

A method of quick cooking pasta products and a method used for their manufacture have been patented (Oh *et al.*, 2000) in which toasting has been performed under controlled conditions after sheeting or extrusion. Roasting alters the structure of fats, denatures the proteins and gelatinizes the starch to dextrins and then reducing sugars. Losses of amino acids due to roasting is an important factor; the higher the temperature or longer the times, the more losses there are. Severe loss of lysine (88%) and thiamin (30–90%) has been reported for toasting/roasting of breakfast cereals and meat (Fellows, 2009).

Roasting or toasting enhances the shelf-life of cereal products by inactivating the contaminating microorganisms and enzymes present in them. Roasted meat or poultry products have a chance to contain pathogens, and thus are to be stored in chilled or frozen temperatures or modified atmospheric storage (Smith *et al.*, 2004).

Antioxidative activity of green and roasted coffee beans is markedly affected by roasting methods like convection or microwave with or without pre-drying or humidifying (Nebesny and Budryn, 2003). Microwave heating for coffee roasting protects its antioxidative properties better than convectional heating. The caffeine content decreases marginally upon microwaving but a greater loss occur due to convectional roasting. The polyphenols content is lowest in pre-dried, convection roasted beans and highest in beans subjected to combined convection–microwave roasting. The 5-hydroxymethylfurfural concentration is highest in beans dried before roasting and less is formed upon microwave heating than upon convectional heating.

10.6 Recent researches

Product quality from chickpea (*Cicer arietinum* L.) changes when the *dhal* (split pulse) or flour is subjected to toasting. *Dhal* and flour when toasted show differences in their odour profiles and changes in the protein characteristics by decreasing the high molecular weight protein fraction, the sensory profile and the quality of the fried product (Ravi, Ajila and Prasada Rao, 2011).

When toasted at different times and temperatures, the moisture content decrease of bread slices follows an exponential trend (Capuano *et al.*, 2009). During toasting, browning is more intense when toasting is conducted at higher temperatures, and linear correlations exist between browning, 5-hydroxymethyl-2-furaldehyde (HMF) and acrylamide concentration when toasted at 180 °C. The HMF and acrylamide contents increase with the toasting time and temperature. In bread (made of wheat, rye and whole wheat flours) when toasted at different temperatures and times, the rye model system produces more Maillard reaction products such as HMF and acrylamide at all temperatures, while whole wheat systems produce less HMF, but more acrylamide is formed than the wheat sample. Some of the additives like glycine, asparaginase and antioxidant extract from green tea change the acrylamide formation and antioxidant activity. The addition of glycine is effective in reducing acrylamide formation, but increases browning development, antioxidant activity and HMF formation. Asparaginase reduces acrylamide formation up to 88% but shows no effect on browning and antioxidant activity. The addition of antioxidant compounds from green tea produces no clear effect on acrylamide formation (Capuano *et al.*, 2009).

Roasting time and temperature influences the formation of acrylamide in almond. However, a short-term storage at elevated temperature reduces the acrylamide levels in roasted almonds (Zhang *et al.*, 2011). Roasting of buckwheat flour causes a decrease in total phenolics, total flavonoids and antioxidative activities (Zhang *et al.*, 2010). The slow disintegration rate and the high amount of swelling of almonds in the stomach give high satiety in consumption. Roasting significantly improves the disintegration rates of almonds and increases loss of solids during simulated gastric digestion (Kong and Singh, 2009). Roasting shows more desirable effects on the amino acid content of the fish samples; for example roasting increases the total amino acid contents of four marine fishes commonly consumed in Nigeria (Oluwaniyi, Dosumu and Awolola, 2010). Sequential infrared and hot air (SIRHA) roasting of almond has been claimed to have more bacterial reductions compared to hot-air roasting (Yang *et al.*, 2010). The effects of three traditional processing methods such as fermentation, roasting and germination have been investigated on the nutritional composition of bambara groundnut (BG) seeds (Ijarotimi and Esho, 2009). The total amino acid contents and relative proportions of essential amino acids are higher

in germinated BG seed meal than in fermented and roasted BG meals. The antinutritional factors in fermented meal are low and protein levels are higher compared to those in germinated and roasted meals (Ijarotimi and Esho, 2009).

The toasting process has been reported to increase the contents of resistance starch and insoluble dietary fibre and antioxidant properties of pasta enriched with chickpea flour (Fares and Menga, 2012). The addition of chickpea flour during pasta preparation from durum wheat increases the total phenolic content and total free phenolic acid content in the uncooked pasta, whereas in the cooked pasta, the bound phenolics fraction increases due to the presence of durum wheat.

10.7 Possible future applications

Roasting or toasting is a common, popular and cheap unit operation in food industries. Research organizations and industries are constantly trying to improve the product quality with a simultaneous decrease in energy expenditure, better roaster/toaster design and by using the on-line temperature–time controlling device. The other areas of research include development of new generation breakfast cereals and snacks with unique quality attributes and application of alternative fuels and processing methods by designing energy-efficient toasting machines. Microstructural observations and the imaging technique can also help in understanding the functional behaviour of the developed products.

10.8 Conclusions

Roasting or toasting is a major unit operation in the breakfast cereals or snack industry in terms of value addition. The process of roasting or toasting induces many changes in the food materials, including the development of a crisp texture and characteristic flavour, which are the important characteristics of this operation. The effect of temperature, time and moisture content markedly influences the quality of the roasted or toasted food. The availability of cost-effective machinery is still a constraint for their wide use in other industries apart from snack and breakfast cereal manufacturing. There exists a research need to understand the relationship between product attributes, consumer acceptability, structure and processing methods employing roasting or toasting processes.

Symbols

a	Food granule surface area per unit packed bed volume, m^2/m^3
a, b	Constants
C_p	Specific heat, J/kg K
D	Component mass diffusivity, m^2/s
G	Component mass velocity, kg/s m^2
$h_{\text{conv mass}}$	Convective mass transfer coefficient, m/s
h_{conv}	Convective heat transfer coefficient, W/m^2 K
h	Specific enthalpy, J/kg
I	Characteristic transfer dimension, m
k	Thermal conductivity, W/m K
N_{Sch}	Schmidt number
N_{Pr}	Prandtl number
N_{Re}	Reynolds number
N_{Nu}	Nusselt number
N_{BiH}	Biot number for heat transfer
N_{BiM}	Biot number for mass transfer
N_T	Decimal fraction thermal efficiency
N_W	Maximum drying rate, kg H_2O/s
Q	Component enthalpy flow, W
R_e	Decimal fraction granules bed recirculation air ratio
T	Temperature, K
V	Volume, m^3
W	Liquid phase component mass fraction, kg component/kg total liquid phase
Greek letters	
ρ	Mass density, kg/m^3
Subscripts	
v	Vapour phase at food granule surface
w	Wetted food granule surface

References

Adrian, J. (1982) The Maillard reaction. In: *Handbook of the Nutritive Value of Processed Food* (M. Rechcigl, ed.), vol. **1**. CRC Press, Boca Raton, Florida, USA, pp. 529–608.

Adrian, L.D., Ann, L.D. and Marcus, L.V. (2000) Breakfast cereal biscuit comprising waxy grain. Patent WO 2000028836 A1.

Akinoso, R. and Raji, A.O. (2011) Optimization of oil extraction from locust bean using response surface methodology. *European Journal of Lipid Science and Technology*, **113**(2), 245–252.

Amoo, I.A. (2004) Effect of roasting on the chemical composition of coconut (*Cocos nucifera*) seed flour and oil. *Journal of Food, Agriculture and Environment*, **2**, 18–20.

Aremu, M.O., Olaofe, O., Basu, S.K., Abdulazeez, G. and Acharya, S.N. (2010) Processed cranberry bean (*Phaseolus coccineus* L.) seed flour for the African diet. *Canadian Journal of Plant Science*, **90**(5), 719–728.

Ari, M.M., Ayanwale, B.A., Adama, T.Z. and Olatunji, E.A. (2012) Evaluation of the chemical composition and antinutritional factors (ANFs) levels of different thermally processed soybeans. *Asian Journal of Agricultural Research,* **6**(2), 91–98.

Bhat, R. and Sridhar, K.R. (2011) Influence of ionizing radiation and conventional food processing treatments on the status of free radicals in lotus seeds: an ESR study. *Journal of Food Composition and Analysis*, **24**(4–5), 563–567.

Bhat, R., Sridhar, K.R. and Bhushan, B. (2007) Free radicals in velvet bean seeds (*Mucuna pruriens* L. DC) and their status after gamma-irradiation and conventional processing. *LWT-Food Science and Technology*, **40**(9), 1570–1577.

Bhattacharya, S. and Prakash, M. (1997) Kinetics of roasting of split chickpea (*Cicer arietinum*). *International Journal of Food Science and Technology*, **32**, 81–84.

Bialobrzewski, I., Danowska-Oziewics, M., Karpinska-Tymoszczyk, M., Nalepa, B., Markowski, M. and Myhan, R. (2009) Turkey breast roasting-process optimization. *Journal of Food Engineering*, **96**(3), 394–400.

Boateng, J., Verghese, M. Walker, L.T. and Ogutu, S. (2008) Effect of processing on antioxidant contents in selected dry beans (*Phaseolus* spp. L.). *LWT-Food Science and Technology*, **41**(9), 1541–1547.

Broncano, J.M., Petron, M.J., Parra, V. and Timon, M.L. (2009) Effect of different cooking methods on lipid oxidation and formation of free cholesterol oxidation products (COPs) in Latissimus dorsi muscle of Iberian pigs. *Meat Science*, **83**(3), 431–437.

Capuano, E., Ferrigno, A., Acampa, I., Serpen, A., Acar, O.C., Gokmen, V. and Fogliano, V. (2009) Effect of flour type on Maillard reaction and acrylamide formation during toasting of bread crisp model systems and mitigation strategies. *Food Research International*, **42**(9), 1295–1302.

Chen, G., Song, H. and Ma, C. (2009) Aroma-active compounds of Beijing roast duck. *Flavour and Fragrance Journal*, **24**(4), 186–191.

Davidson, V.J., Brown, R.B. and Landman, J. J. (1999) Fuzzy control system for peanut roasting. *Journal of Food Engineering*, **41**, 141–146.

Demir, A.D., Baucour, P., Cronin, K. and Abodayeh, K. (2003) Analysis of temperature variability during the thermal processing of hazelnuts. *Innovative Food Science and Emerging Technologies*, **4**(1), 69–84.

Eichner, J. (1999) Coffee roasting methods. US Patent 6,607,768 B1.

Ewald, H.T. and Simmons, P.G. (2011) Steam and platen toasting device. US Patent 2011/0256286 A1.

Fagbemi, T.N. (2008) The influence of processing techniques on the energy, ash properties and elemental composition of cashew nut (*Anacardium occidentale* Linn). *Nutrition and Food Science*, **38**(2), 136–145.

Fares, C. and Menga, V. (2012) Effects of toasting on the carbohydrate profile and antioxidant properties of chickpea (*Cicer arietinum* L.) flour added to durum wheat pasta. *Food Chemistry*, **131**(4), 1140–1148.

Fast, R.B. (1990) Manufacturing technology of ready-to-eat cereals. In *Breakfast Cereals and How They Are Made* (Fast, R.B. and Caldwell, E.F., eds). American Association of Cereal Chemists, St Paul, Minnesota, USA, pp. 15–42.

Fellows, P.J. 2009 *Food processing technology: Principles and practice*. 3rd edn CRC press Boca Raton and Woodhead Publishing Ltd, Cambridge, New Delhi, pp 913.

Goni, S.M. and Salvadori, V.O. (2010) Prediction of cooking times and weight losses during meat roasting. *Journal of Food Engineering*, **100** (1), 1–11.

Goni, S.M. and Salvadori, V.O. (2012) Model based multi-objective optimization of beef roasting. *Journal of Food Engineering*, **111**(1), 92–101.

Hecimovic, I.., Belscak-Cvitanovic, A., Horzic, D. and Komes, D. (2011) Comparative study of polyphenols and caffeine in different coffee varieties affected by the degree of roasting. *Food Chemistry*, **129**(3), 991–1000.

Ijarotimi, O.S. and Esho, T.R. (2009) Comparison of nutritional composition and anti-nutrient status of fermented, germinated and roasted bambara groundnut seeds (*Vigna subterranea*). *British Food Journal*, **111**(4), 376–386.

Kemper, T.G. (2000) Innovations in meal desolventizing. *Inform – Champaign*, **11**(9), 967, 969–972.

Kidmose, U., Christensen, L.P., Agili, S.M. and Thilsted, S.H. (2007) Effect of home preparation practices on the content of provitamin A carotenoids in coloured sweet potato varieties (*Ipomoea batatas* Lam.) from Kenya. *Innovative Food Science and Emerging Technologies*, **8**(3), 399–406.

Kirbaslar, F.G. and Erkmen, G. (2003) Investigation of the effect of roasting temperature on the nutritive value of hazelnuts. *Plant Foods for Human Nutrition*, **8**, 1–10.

Kunyanga, C.N., Imungi, J.K., Okoth, M.W., Biesalski, H.K. and Vadivel, V. (2011) Flavonoid content in ethanolic extracts of selected raw and traditionally processed indigenous foods consumed by vulnerable groups of Kenya: antioxidant and type II diabetes-related functional properties. *International Journal of Food Sciences and Nutrition*, **62**(5), 465–473.

Kong, F. and Singh, R.P. (2009) Digestion of raw and roasted almonds in simulated gastric environment. *Food Biophysics*, **4**(4), 365–377.

Liao, G.Z., Wang, G.Y., Xu, X.L. and Zhou, G.H. (2010) Effect of cooking methods on the formation of heterocyclic aromatic amines in chicken and duck breast. *Meat Science*, **85**(1), 149–154.

McGorrin, R.J. (2006) Character-impact flavour compounds. In: *Sensor Directed Flavour Analysis* (Marsili, R., ed.). CRC Press, pp. 223–267.

Manay, N.S. and Shadaksharaswamy, M. (2008) *Foods: Facts and Principles*. 3rd edn. New Age International (P) Ltd, Publishers, New Delhi, India, p. 467.

Nazzaro, M., Barbarisi, C., La Cara, F., Volpe, M.G. (2011) Chemical and biochemical characterisation of an IGP ecotype chestnut subjected to different treatments. *Food Chemistry*, **128**(4), 930–936.

Nebesny, E. and Budryn, G. (2003) Antioxidative activity of green and roasted coffee beans as influenced by convection and microwave roasting methods and content of certain compounds. *European Food Research and Technology*, **217**(2), 157–163.

Oh, N.H., Tecedor, S.L., Schryer, R.F. and Meyers, E.J. (2000) Wet toasted pasta and method of manufacture. US Patent **6**,106,882.

Oluwaniyi, O.O., Dosumu, O.O. and Awolola, G.V. (2010) Effect of local processing methods (boiling, frying and roasting) on the amino acid composition of four marine fishes commonly consumed in Nigeria. *Food Chemistry*, **123**(4), 1000–1006.

Potter, N.N. and Hotchkiss, J.H. (1995) *Food Science*, 5th edn. New York: Chapman & Hall, New York, USA, 608 pp.

Pradeep, S.R. and Guha, M. (2011) Effect of processing methods on the nutraceutical and antioxidant properties of little millet (*Panicum sumatrense*) extracts. *Food Chemistry*, **126**(4), 1643–1647.

Ravi, R., Ajila, C.M., Prasada Rao, U.J.S. (2011) Role of steaming and toasting on the odor, protein characteristics of chickpea (*Cicer arietinum* L.) flour, and product quality. *Journal of Food Science*, **76**(2), S148–S155.

Rocha, S., Maeztu, L., Barros, A., Cid, C. and Coimbra, M.A. (2003) Screening and distinction of coffee brews based on headspace solid phase microextraction/gas chromatography/principal component analysis. *Journal of the Science of Food and Agriculture*, **84**, 43–51.

Scher, L.I., Fazio, P. and Hsieh, M.W. (1993) Process design and analysis of dry cooking operations. *AICHE Symposium Series, New Jersey*, **89**(297), 1–31.

Shen, C., Geornaras, I., Belk, K.E., Smith, G.C. and Sofos, J.N. (2011) Inactivation of *Escherichia coli* O157:H7 in moisture-enhanced non intact beef by pan-broiling or roasting with various cooking appliances set at different temperatures. *Journal of Food Science*, **76**(1), M64–M71.

Schenker, S., Heinemann, C., Huber, M., Pompizzi, P., Perren, R. and Escher, F. (2002) Impact of roasting conditions on the formation of aroma compounds in coffee beans. *Journal of Food Science*, **67**(1), 60–66.

Smith, J.P., Daifas, D.P., El-Khoury, W., Koukoutsis, J. and El-Khoury, A.A. (2004) Shelf life and safety concerns of bakery products – a review. *Critical Reviews in Food Science and Nutrition*, **44**, 19–56.

Sumati, R.M., Shalini, M.R. and Rajagopal, M.V. (2006) *Food Science*, 2nd edn. New Age International Publisher, New Delhi, 226 pp.

Sumithra, B. and Bhattacharya, S. (2008) Toasting of corn flakes: product characteristics as a function of processing conditions. *Journal of Food Engineering*, **88**(3), 419–428.

Toledo, T. and Brody, A.L. (1999) Food science, technology and engineering overview for food product development. In: *Developing New Food Products for a Changing Market Place*. (Brody, A.L. and Lord, J.B., eds). CRC Press, Inc., USA.

Uysal, N., Sumnu, G. and Sahin, S. (2009) Optimization of microwave-infrared roasting of hazelnut. *Journal of Food Engineering*, **90**(2), 255–261.

Veltrop, L. and D'van, E. (2011) Bread product edge toasting shield. US Patent 2011/0059210 A1.

Wang, N., Fu, Y. and Lim, L.T. (2011) Feasibility study on chemometric discrimination of roasted Arabica coffees by solvent extraction and Fourier transform infrared spectroscopy. *Journal of Agricultural and Food Chemistry*, **59**(7), 3220–3226.

Wattanachant, S., Benjakul, S. and Ledward, D.A. (2005) Effect of heat treatment on changes in texture, structure and properties of Thai indigenous chicken muscle. *Food Chemistry*, **93**(2), 337–348.

Yang, J., Bingol, G., Pan, Z., Brandl, M.T., McHugh, T.H. and Wang, H. (2010) Infrared heating for dry-roasting and pasteurization of almonds. *Journal of Food Engineering*, **101**(3), 273–280.

Zhang, M., Chen, H., Li, J., Pei, Y. and Liang, Y. (2010) Antioxidant properties of tartary buckwheat extracts as affected by different thermal processing methods. *LWT-Food Science and Technology*, **43**(1), 181–185.

Zhang, G., Huang, G., Xiao, L., Seiber, J. and Mitchell, A.E. (2011) Acrylamide formation in almonds (*Prunus dulcis*): influences of roasting time and temperature, precursors, varietal selection, and storage. *Journal of Agricultural and Food Chemistry*, **59**(15), 8225–8232.

11

Micronization and Encapsulation: Application of Supercritical Fluids in Water Removal

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11.1 Introduction

Encapsulation is defined as the process of forming a thin shell over a solid, liquid or gaseous material, which is completely contained within the capsule wall, and micronization is defined as the process of reducing the particle size and obtaining micro- or even nano-sized particles (Cocero *et al.*, 2009).

Production of ultrafine (micro- or nano-sized) particles with desired properties and precise control of particle size and morphology is one of the objectives of many industries. Conventional processes for particle formation suffer from limitations in producing a desirable final product. The use of supercritical fluids for particle sizing and design represents an attractive alternative to create micro- and nano-sized particles with controlled particle size and narrow particle size distribution. Furthermore, these processes offer a wide control of particle morphology, obtain solvent-free products and avoid thermal degradations due to the low level of the operating temperatures (since in most cases carbon dioxide is the supercritical fluid) (Cocero *et al.*, 2009; Gomes *et al.*, 2012).

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Many different processes for particle formation using supercritical fluids have been proposed; in each one, the supercritical fluids perform different functions, such as solvent (rapid expansion of supercritical solutions (RESS)), antisolvent (supercritical antisolvent (SAS) precipitation), co-solvent or solute (particles from gas-saturated solutions (PGSS)) and propellant (carbon dioxide-assisted nebulization with a bubble dryer (CAN-BD)) (Martín *et al.*, 2010a). Due to the different process arrangements and apparatuses used in these particle formation processes, different acronyms are also used by the various authors to promote the uniqueness of the process, despite not being very different one from the other.

In particular, the methods referred to as CAN-BD and PGSS drying allow the production of ultrafine particles of water-soluble compounds. These processes micronize or encapsulate the desired compound or compounds by removing the water. The main target of them is to obtain stabilized dry powders. These technologies are promising when applied to proteins or other biomolecules that may be denatured by conventional drying processes like spray drying. Different from freeze drying, it is also possible to control the particle size and particle size distribution using these processes (Perrut, 2004). Furthermore, the time required for drying by freeze drying is much longer (hours rather than seconds) and there is no need for additional milling or micronization steps (Sievers *et al.*, 2001). Spray drying and freeze drying are two of the most successful conventional methods used to produce particles by removing water. Table 11.1 shows the main limitations of both techniques.

Table 11.1 Limitations of spray drying and freeze drying methods

Conventional method	Principle of operation	Limitation	Reference
Spray drying	Atomization with a nozzle or spinning wheel of a mixture (core and carrier materials) into a hot-air desiccant into a chamber	Problems with efficient particle collection and the potential instability of materials sensitive to high temperatures	Prinn, Costantino and Tracy (2002); Martín *et al.* (2010a); Santos and Meireles (2010)
Freeze drying	Water is removed from the frozen state by vacuum sublimation, maintaining the drying chamber pressure and temperature below the triple point of water	Long processing time, expensive process costs and difficult to control the particle size	Perrut (2004); Martín *et al.* (2010a); Santos and Meireles (2010)

The present chapter describes the basic aspects of CAN-BD and PGSS drying processes for the production of ultrafine particles. For each technique, the history, the process description and the influence of process variables have been discussed. Recent developments focused on processing of food ingredients using these methods are also presented.

11.2 Supercritical fluid

A supercritical fluid is defined as a fluid whose pressure and temperature are simultaneously higher than those at the critical point (Figure 11.1). Its solvency power is enhanced due to its higher density, which is very similar to those of liquids (100–900 kg/m^3 at 7.5–50 MPa). Furthermore, these fluids have the main characteristics of gases such as low viscosity, large diffusivity and small surface tension, which are favourable attributes for several processes (Skala and Orlovic, 2006). Carbon dioxide is a fluid widely applied to produce fine particles using supercritical fluid methods. The main advantages of CO_2 are its nontoxicity, nonflammability, its relatively low critical temperature (T_C = 304.2 K) and pressure (P_C = 7.38 MPa), it is inexpensive and recyclable. In addition, it can be completely separated from the final product by expansion and then liquefied for recycling purpose.

11.3 Developmental stages

Carbon dioxide-assisted nebulization with a bubble dryer (CAN-BD) was developed based on the same concept of the PGSS technique and patented

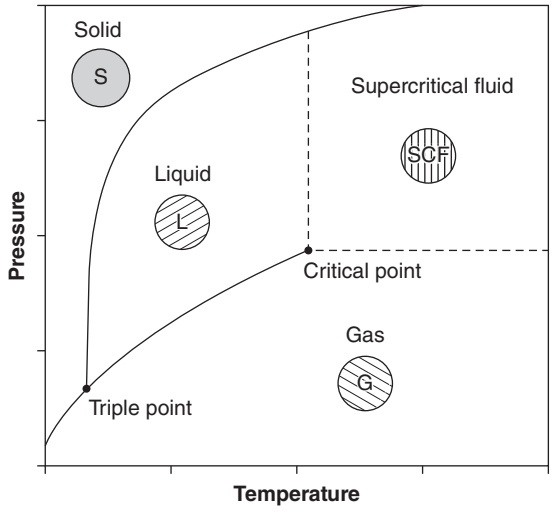

Figure 11.1 Pressure–temperature phase diagram of a single substance

by Sievers and co-author in 1997 (Sievers and Karst, 1997). These authors proposed a modification to the PGSS process that allowed expanding the process application for the use of any compound that is water soluble (Nunes and Duarte, 2011). In the PGSS concept, a gas-saturated solution is expanded through a nozzle to an atmospheric pressure. During the expansion, the gas dissolved into the solution is suddenly vaporized and the intense cooling due to the Joule–Thomson effect during CO_2 expansion promotes particle formation (Martín and Weidner, 2010). In the CAN-BD process, an aqueous solution mixed with CO_2 is expanded to atmospheric pressure through a flow restrictor to generate aerosols of microbubbles and microdroplets. This process is available commercially (Sievers, Best and Cape, 2008). Since the first patent in 1997, several modifications were proposed in order to make the CAN-BD process more versatile; these variants are referred to as supercritical-assisted atomization (SAA) and supercritical enhanced atomization (SEA), proposed by Padrela *et al.* (2010) and Reverchon (2002), respectively.

Another modification of the PGSS process is PGSS drying. The main difference between the two techniques is that in PGSS drying, the coating material is fed to the static mixer in an aqueous solution (Martín *et al.*, 2010a). This process was patented by Weidner and co-workers in 2000 (Weidner *et al.*, 2000). In this technique, the particles do not need to be dried with the help of N_2 inert flux as in CAN-BD. Although the published papers mostly focus on drugs and polymers, some authors demonstrated that this technique is also feasible for food applications (Weidner, 2009). This process exists at the pilot and industrial scale, providing a basis for the demonstration of its technical and economic feasibility for some industrial applications (Nunes and Duarte, 2011; Weidner 2009).

11.4 Process description and influence of process parameters

11.4.1 CAN-BD

Process description The CAN-BD process consists of mixing a stream of compressed CO_2 with an aqueous solution or dispersion containing the solute of interest during a short time in a 'low-dead-volume' tee (< 1 μL). A quantity of CO_2 is solubilized in water and an emulsion is formed. Carbon dioxide is a suitable compressible fluid because it has a good solubility in water (1.6 mole% at 336 K and 10 MPa) and its use enhances the expansion process. The emulsion so formed is rapidly decompressed to atmospheric pressure through a flow restrictor (or a capillary tube) by expansion into a drying chamber that generates an aerosol of very fine droplets of liquid-carrying microbubbles of gas (Sievers *et al.*, 1999). This plume of microbubbles and

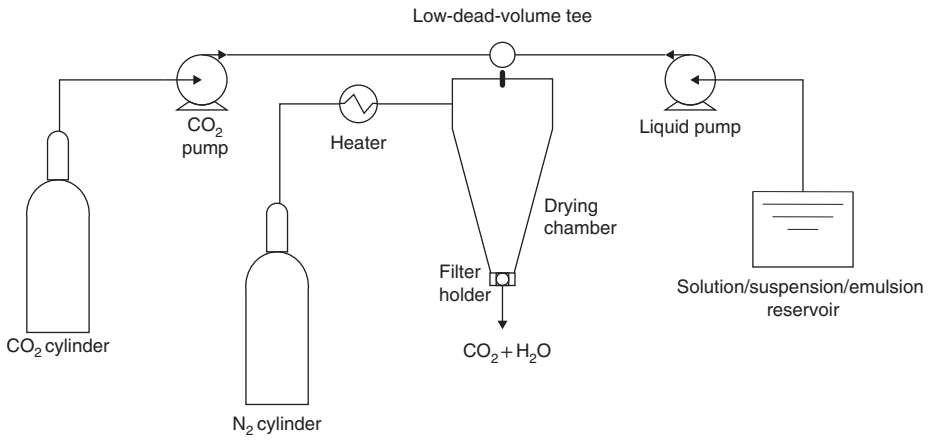

Figure 11.2 Schematic diagram of the carbon dioxide-assisted nebulization with a bubble dryer (CAN-BD) process

microdroplets is further dried by contact with a stream of inert N_2 warmed at temperatures between 298 and 348 K, much lower than those currently used in spray drying (Perrut, 2004). CAN-BD has been used with aqueous and organic solvents. Figure 11.2 represents a schematic diagram of the CAN-BD process.

There are two versions of this process, static and dynamic. The static version involves the pre-mixing of the supercritical CO_2 and the aqueous solution, and after the equilibrium is approached, the mixture in a high pressure chamber is allowed to expand to atmospheric pressure. The dynamic version involves continuous intimate mixing of the aqueous solution and the supercritical CO_2 (Cape *et al.*, 2008).

The CAN-BD process provides a more efficient atomization because, after the primary atomization, the release of CO_2 produces a secondary atomization, producing smaller droplets. This technique does not differ much from the classical aerosol method. Due to the very short contact time between the compressible CO_2 and the aqueous solution, the CO_2 is far from dissolving to saturation in the aqueous solution, unlike the PGSS and PGSS drying. It is noteworthy that owing to the depressurization of the CO_2, a sharp temperature decrease takes place (the Joule–Thomson effect) both in the tee and in the restrictor, which also must be heated to avoid plugging. The temperature required can damage some thermolabile compounds, which can be solved by applying a vacuum in the drying chamber (Tabernero, Valle and Galán, 2012; Charbit, Badens and Boutin, 2004).

Influence of process parameters The CAN-BD process has been developed mainly for industrial purposes. Therefore, there are not many published papers that investigated the influence of the operational parameters on particle characteristics. In a first study, Sievers *et al.* (1999) produced fine

particles of pharmaceuticals and other materials by aerosolization in a low-volume mixing device. The authors observed that the desired fine aerosols are obtained when a 'low-dead-volume' tee (< 1 μL) was used, but under the same conditions, using a normal 1/16 in (0.159 cm) tee with a volume of about 50 μL, no aerosol was obtained. Furthermore, a water stream rather than an aerosol resulted when the CO_2 tee inlet pressure was set at 5.5 MPa instead of at 10.3 MPa.

Sievers *et al.* (1999) also noted that the size distribution of the particles suspended in the formed aerosol depended on the concentration of the precursors in the aqueous solution. Increasing the concentration of the precursors a narrower size range was obtained. The results of process parametric studies for ethanolic solutions containing beta-methasone as well as aqueous solutions containing mannitol or myo-inositol indicated that the ratio of CO_2 to the solution mass flow rate has a significant influence on the particle size. By manipulating this ratio, the particle size of fine powders generated by the CAN-BD process can be controlled from nanometre size to micrometre size (Huang *et al.*, 2003).

According to Sievers *et al.* (2000), spherical morphology might be expected for particles formed by drying microbubbles or droplets. Liquid droplets synthesized by aerosol methods maintain a spherical shape to minimize surface energy. However, these authors observed that differences in drying conditions can lead to different morphologies of particles of the same drug. Lifeboat-shaped particles of cromolyn sodium were obtained when the aerosol plume was bubble-dried using the dynamic method by mixing with dry nitrogen at temperatures between 348 and 368 K. Nonetheless, when cromolyn sodium was dried at or near room temperature by passing the aerosol plume over the concentrated sulfuric acid, more nearly spherical particles were observed. The concentrated sulfuric acid was alternatively used as a desiccant to dry the aerosol plume diluted with nitrogen before collection on a filter.

Applications The CAN-BD process has been applied to obtain powders from water solutions of pharmaceuticals (Sievers *et al.*, 2007), proteins (Cape *et al.*, 2008) and other water-soluble compounds. Table 11.2 summarizes the various substances that have been processed into particles using the CAN-BD technique.

Despite the potential of CAN-BD to dry water-soluble compounds for food applications, data for this process are scarcely available in the literature. Dried powders of extracts from natural sources using the CAN-BD technique have been obtained (Andersson *et al.*, 2012; Herrero *et al.*, 2010) and the process was patented in 2009 as an on-line process in one step combining the pressurized hot water extraction (PHWE) and the drying of the extract by the CAN-BD process, named as water extraction and particle formation on-line (WEPO) (Figure 11.3) (Ibáñez *et al.*, 2009). This promising process

Table 11.2 Summary of particles produced via carbon dioxide-assisted nebulization with a bubble dryer (CAN-BD) process

Substance	Solvent/nebulization fluid	Operational conditions	Results	References
Enzyme α1-antitrypsin	Water/CO_2	Mixing tee pressure: 8.27 MPa Mixing tee temperature: room temperature Solution flow rate: 0.3–0.5 mL/min N_2 drying gas flow rate: 15–30 L/min Drying temperature: 313 K	Particle formation type: micronization Morphology: spherical Moisture content: 1.8% Particle size: 1.9 — 2.2 μm	Cape *et al.* (2008)
Trypsinogen (protein)	Water/CO_2	Mixing tee pressure: 8.27 MPa Mixing tee temperature: room temperature Solution flow rate: 0.3–0.5 mL/min N_2 drying gas flow rate: 15–30 L/min	Particle formation type: micronization Morphology: dimpled raisin-like Particle size: 0.86–1.4 μm	Cape *et al.* (2008)
NaCl	Water/CO_2	Mixing tee pressure: 8 MPa Mixing tee temperature: room temperature Solution flow rate: 0.3 mL/min Nebulizing fluid flow rate: 1–3 mL/min N_2 drying gas flow rate: 15 L/min Drying temperature: 333–353 K	Particle formation type: micronization Morphology: hollow clusters Particle size: 2–5 μm	Sievers *et al.* (2003)
Palmitic acid	Ethanol/CO_2	Mixing tee pressure: 8 MPa Mixing tee temperature: room temperature Solution flow rate: 0.3 mL/min Nebulizing fluid flow rate: 1–3 mL/min N_2 drying gas flow rate: 15 L/min Drying temperature: 333–353 K	Particle formation type: micronization Morphology: flat, leaf-shaped Particle size: 1 μm	Sievers *et al.* (2003)

Table 11.2 (*continued*)

Substance	Solvent/nebulization fluid	Operational conditions	Results	References
Lysozyme and lactate dehydrogenase	Water/CO_2	Mixing tee pressure: 10.34 MPa Mixing tee temperature: >305 K Solution flow rate: 0.3 mL/min N_2 drying gas flow rate: 15 L/min Drying temperature: <343 K	Particle formation type: micronization Morphology: spherical Moisture content: <4% Particle size: 1–3 μm	Sellers *et al.* (2001)
Onion extract	Water/CO_2	Mixing tee pressure: 8 MPa Mixing tee temperature: 393 K Solution flow rate: 0.2–0.3 mL/min Nebulizing fluid flow rate: 3–10 mL/min Drying temperature: 393 K	Particle formation type: micronization Morphology: spherical Moisture content: 4 % Particle size: < 4 μm	Andersson *et al.* (2012)
Rosemary extract	Water/CO_2	Mixing tee pressure: 8 MPa Mixing tee temperature: 473 K Solution flow rate: 0.2 mL/min Drying temperature: 343 K	Particle formation type: micronization	Herrero *et al.* (2010)

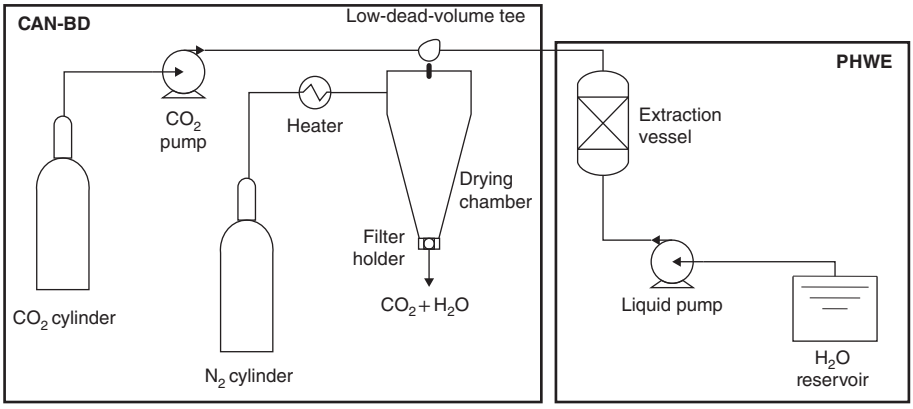

Figure 11.3 Schematic diagram of the WEPO process

was suitable to obtain fine powder with particle sizes smaller than 4 μm in diameter with an intact antioxidant capacity of great interest for food and cosmetic industries. Andersson *et al.* (2012) compared the particles obtained by WEPO with the ones obtained by PHWE followed by freeze drying. Despite having similar results in terms of antioxidant capacity, concentration of quercetin derivatives and water content, the WEPO process was able to produce smaller and well-defined spherical particles. According to Herrero *et al.* (2010), the results obtained via WEPO are promising considering the time saving due to the absence of a later drying process.

11.4.2 PGSS drying

Process description The PGSS drying process (Figure 11.4) consists of mixing an aqueous solution with supercritical CO_2 using a static mixer at the desired temperature and pressure. Aqueous suspensions (Paz, Martín and Cocero, 2012) and oil-in-water emulsions (Varona *et al.*, 2010) also can be fed to the static mixer. In this mixer, a certain amount of water is extracted by CO_2 despite supercritical CO_2 being partly dissolved in the liquid solution. This biphasic mixture is expanded down to atmospheric conditions through a nozzle into a thermally insulated spray tower (Varona *et al.*, 2010). The expansion of the CO_2 dissolved into the liquid promotes the formation of fine droplets. In addition, evaporation of the remaining water takes place due to pre-selected temperature conditions in the spray tower. Water is exhausted together with CO_2 by a blower (supercompressor) from the spray tower. In order to achieve a good water and CO_2 removal from the powder, the conditions in the spray tower have to be above the dew line of the water–CO_2 mixture because in this region one single gas phase is obtained. Knowledge of the vapour–liquid equilibrium (VLE) diagram of this mixture is needed

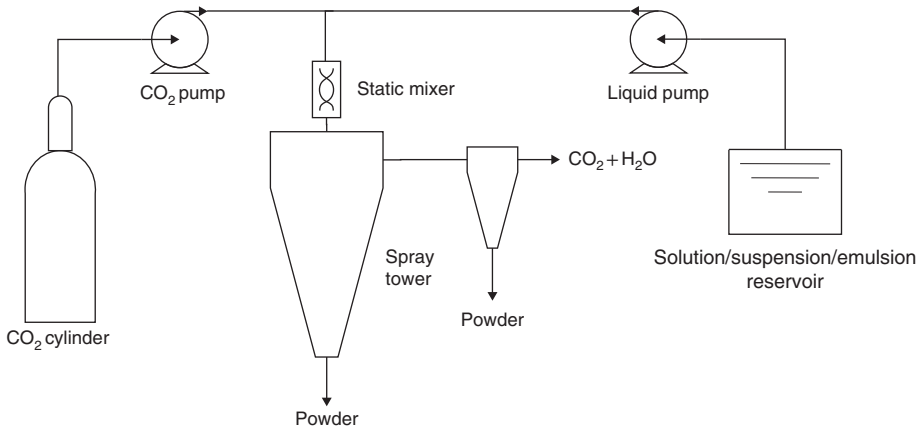

Figure 11.4 Schematic diagram of the PGSS drying process

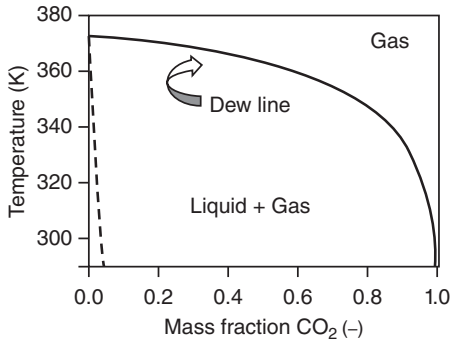

Figure 11.5 Temperature–composition diagram for water and carbon dioxide at atmospheric pressure

to choose between different strategies for water removal. For instance, the temperature in the spray tower can be increased and therefore less CO_2 is used; the other option is to work at a lower temperature and thus it is necessary to increase the mass flow of CO_2 to remove the same amount of water (Figure 11.5) (Pham, Pollak and Petermann, 2012; Martín and Weidner, 2010; Meterc, Petermann and Weidner, 2008). Finally, the produced powder is collected inside the spray tower with a cyclone. Particles are collected at the bottom of the spray tower and CO_2 together with evaporated water leave the tower through its upper part. A cyclone separator is used to recover fine powder entrapped in the effluent gas (Martín *et al.*, 2010b). This process provides an inert atmosphere (CO_2 atmosphere without oxygen), avoiding the possibility of oxidation (Tabernero, Valle and Galán, 2012).

Influence of process variables The literature provides a detailed experimental analysis of the influence of different process and design variables

and parameters (temperature, pressure, flow rates, design of the static mixer, concentration of carrier material, etc.) on particle size, residual moisture, encapsulation efficiency and morphology of the particles obtained by PGSS drying.

Particle size Experimental results demonstrated that one of the major parameters influencing the particle size is the ratio of the gas to solution flow rate. The particle size decreases as the ratio of gas to solution flow rate increases, due to a more efficient atomization and faster precipitation (Martín *et al.*, 2010b). A reduction of particle size was also observed by Martín *et al.* (2010b) when the number of mixing elements was increased from 0 to 5. The authors concluded that with five mixing elements, a saturation of the liquid phase with CO_2 was achieved. The use of mixing elements favours the contact between the aqueous and gas phases, leading to a more efficient supercritical fluid dissolution into the liquid phase, and improves atomization and particle formation in the spray tower. Varona, Martín and Cocero (2011) observed that the higher carrier material concentration has made the atomization more difficult; this is due to the higher emulsion viscosity which produced bigger particles.

Pre-expansion temperature and pressure are important parameters to determine the extraction conditions in the static mixer. These parameters have to be chosen in order to promote a higher concentration of CO_2 in the gas-saturated solution, which enhances the atomization process and leads to the formation of smaller particles (Paz, Martín and Cocero, 2012).

Residual moisture The main parameters that influence the residual moisture are the ratio of the gas to liquid flow rate, pre-expansion temperature and pressure. The decrease of residual moisture with the ratio of the gas to liquid flow rate has a direct consequence for the mass balance (Martín *et al.*, 2010b). Martín and Weidner (2010) showed that a mass balance can be used to calculate the minimum gas/liquid flow rate required for the complete evaporation of water.

Martín *et al.* (2010b) observed a direct relationship between pre-expansion and post-expansion temperatures. As a result of the conservation of energy during the expansion, when the pre-expansion temperature is higher, the temperature in the spray tower is also higher, being more favourable for water evaporation. On the other hand, the increase of the residual moisture when pre-expansion pressure increases is a consequence of the reduction in the spray tower temperature due to the Joule–Thomson effect.

Morphology Particle morphology is directly related to the residual moisture content of particles. When the water has not been successfully removed,

a paste or gel is obtained instead of a solid powder. When the water concentration is lower, the solid powder normally is produced and consists of spherical particles (Pham, Pollak and Petermann, 2012; Martín *et al.*, 2010b).

Varona *et al.* (2010) observed that two main different particle morphologies like spheres and needles were obtained, depending on the pre-expansion conditions. The generation of spheres is favoured by a high pre-expansion temperature and pressure.

Encapsulation efficiency The influence of different parameters on encapsulation efficiency has been investigated by Varona *et al.* (2011, 2010) and Paz, Martín and Cocero (2012). When the pre-expansion temperature is increased, more water is extracted in the static mixer. With a concentrated solution already formed in the static mixer, particles can more easily be surrounded by a shell of carrier material that can be maintained upon drying in the spray tower, leading to the production of microcapsules and an increase in the encapsulation efficiency (Paz, Martín and Cocero, 2012; Varona, Martín and Cocero, 2011).

The observations of Varona, Martín and Cocero (2011) and Paz, Martín and Cocero (2012) diverge with respect to the influence of the concentration of the carrier material on encapsulation efficiency. This divergence may be associated with the different characteristics between the encapsulated materials in the two cases (liquid oil droplets and solid particles).

The pre-expansion pressure and the ratio of gas to product flow rate have a smaller effect on lavandin essential oil encapsulation in modified starch in the conditions investigated by Varona *et al.* (2010). However, in another work, Varona, Martín and Cocero (2011) observed a reduction of lavandin essential oil encapsulation in soybean lecithin when the ratio of gas to product flow rate was increased due to a partial extraction of the oil by supercritical CO_2. However, this effect was not observed by Paz, Martín and Cocero (2012) because the solubility of β-carotene in CO_2 in the conditions studied was very low.

Applications The PGSS drying process has been successfully applied to produce fine particles of polymers and natural compounds. Table 11.3 summarizes the compounds processed by the PGSS drying technique.

Published papers have demonstrated that PGSS drying is feasible for food applications. PGSS drying extends the applicability of PGSS to water-soluble compounds. Application of PGSS drying has been investigated for polymers. Polyethylene glycol (PEG) (Martín *et al.*, 2010b) and polyethylene oxide (PEO) (Pham, Pollak and Petermann, 2012) were dried and micronized by this technique. These polymers can be used as a carrier material for developing formulations of food and pharmaceutical compounds. In addition, the literature provides some natural compounds successfully treated by PGSS drying.

Table 11.3 Summary of particles produced from aqueous solutions using PGSS-drying process

Substance	Solvent	Operational conditions	Results	References
Polyethylene glycol	Water	Pre-expansion pressure: 6.1–15.1 MPa Pre-expansion temperature: 353–414 K Post-expansion temperature: 281–325 K Solution flow rate: 0.5–6 kg/h CO_2 flow rate: 26–100.4 kg/h Gas/liquid flow ratio: 6–139	Particle formation type: micronization Morphology: spherical Moisture content: < 1 % Particle size: 10–20 μm	Martín *et al.* (2010b)
Green tea extracts	Water	Pre-expansion pressure: 5.9–10 MPa Pre-expansion temperature: 383–418 K Post-expansion temperature: 306–352 K	Particle formation type: micronization Morphology: spherical Moisture content: 5.95–13.05% Particle size: < 10 μm	Meterc, Petermann and Weidner (2008)
Gelatine	Water	Pre-expansion pressure: 7.5–8.5 MPa Pre-expansion temperature: 413–433 K Post-expansion temperature: 333–343 K Gas/liquid flow ratio: 20–50	Particle formation type: micronization Moisture content: 8–13% Particle size: 300 μm	Reibe *et al.* (2008)
β-Carotene	Water	Pre-expansion pressure: 8–10 MPa Pre-expansion temperature: 373–403 K Post-expansion temperature: 313–353 K Gas/liquid flow ratio: 21–37	Particle formation type: encapsulation Morphology: agglomerated spheres Particle size: 10–500 μm Encapsulation efficiency (carrier material): 30–60% (soybean lecithin)	Paz, Martín and Cocero (2012)
Lavandin essential oil	Water	Pre-expansion pressure: 6-10 MPa Pre-expansion temperature: 377–403 K Gas/liquid flow ratio: 5–35	Particle formation type: encapsulation Morphology: aggregated spheres Particle size: 1.4–25 μm Encapsulation efficiency (carrier material): 6–14.5% (soybean lecithin)	Varona, Martín and Cocero (2011)
Lavandin essential oil	Water	Pre-expansion pressure: 9–12.1 MPa Pre-expansion temperature: 373–404 K Post-expansion temperature: 333–348 K Solution flow rate: 2.4–4.1 kg/h CO_2 flow rate: 72–91 kg/h Gas/liquid flow ratio: 22.4–41.2	Particle formation type: encapsulation Morphology: spheres and needles Moisture content: 5% Particle size: 15–194 μm Encapsulation efficiency (carrier material): 6–55% (starch)	Varona *et al.* (2010)

PGSS drying was reported on drying green tea extracts containing antioxidant polyphenols. Dried and free-flowing powders were obtained without degradation of the active ingredients because of an oxygen-free atmosphere and low drying temperatures required by this technique, making this process very promising for sensitive substances (Meterc, Peterson and Weidner, 2008). It is possible to produce fine and microbiologically stable gelatin powders having a relatively small amount of CO_2. With the spray drying process it is common to dry aqueous solutions with very low amounts (5%, wt) of high molecular mass gelatin because higher viscosities lead to a blockage of the nozzle due to the formation of fibres and filaments. Therefore, the PGSS drying technology allows a dried food ingredient to be achieved with a considerably reduced energy demand (Reibe *et al.*, 2008).

Lavandin oil-loaded microcapsules have been prepared by using a modified starch, which performed the double function of the surfactant and carrier material (Varona *et al.*, 2010). The encapsulation efficiency varied from 6 to 55%. Particle sizes varied between 15 and 194 μm, with a residual moisture content of about 5%, similar to the water content in unprocessed modified starch. Varona, Martín and Cocero (2011) also studied the formulation of emulsions of lavandin essential oil with liposomes, using soybean lecithin as the carrier material. PGSS drying was effective in micronizing soy lecithin, forming spherical aggregated particles. The low efficiency of encapsulation (6–14.5%) thus obtained can be improved by modifying the process conditions in order to increase the solubility of carbon dioxide in the emulsion in the static mixer. β-Carotene was also encapsulated in soybean lecithin using the PGSS drying process. Dried particles of 10–500 μm were obtained with β-carotene encapsulation efficiencies up to 60% (Paz, Martín and Cocero, 2012).

11.5 Conclusions and future perspectives

CAN-BD and PGSS drying processes use supercritical fluid for water removal. These methods are based on the very simple concept of expanding an aqueous solution saturated with a supercritical fluid, preferably CO_2, through a restriction device. Emulsions and suspensions have also been used. Although no studies reported the use of emulsions via CAN-BD, this operation is possible and extends its applicability. Despite the fact that most published papers explored applications related to the pharmaceutical industry, the present chapter shows the feasibility of applying these processes for producing products for the food industry. Further, the possibility of operating on a large scale exists due to the several advantages presented over the conventional process. The reasons are the prevention of product degradation, control of product characteristics and achievement of a product of good quality due to the closed and inert system used. It is also possible

to couple a process such as WEPO, which combines bioactive compounds (food ingredients) extraction to on-line particle formation. It is a highly promising area and is suitable to obtain desired particles with a reduced time of preparation.

Abbreviations

CAN-BD	Carbon dioxide-assisted nebulization with a bubble dryer
PEG	Polyethylene glycol
PGSS	Particles from gas-saturated solutions
PHWE	Pressurized hot water extraction
PEO	Polyethylene oxide
RESS	Rapid expansion of supercritical solutions
SAA	Supercritical-assisted atomization
SAS	Supercritical antisolvent
SEA	Supercritical enhanced atomization
VLE	Vapour-liquid equilibrium
WEPO	Water extraction and particle formation on-line

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References

Andersson, J.M., Lindahl, S., Turner, C. and Rodriguez-Meizoso, I. (2012) Pressurised hot water extraction with on-line particle formation by supercritical fluid technology. *Food Chemistry*, **134**, 1724–1731.

Cape, S.P., Villa, J.A., Huang, E.T.S., Yang, T.-H., Carpenter, J.F. and Sievers, R.E. (2008) Preparation of active proteins, vaccines and pharmaceuticals as fine powders using supercritical or near-critical fluids. *Pharmaceutical Research*, **25**, 1967–1990.

Charbit, G., Badens, E. and Boutin, O. (2004) Methods of particle production. In: York, P., Kompella, U.B. and Shekunov, B.Y. (eds), *Supercritical Fluid Technology for Drug Product Development*. Informa Healthcare, Marcel Dekker, Aix-en-Provence, France.

Cocero, M.J., Martín, A., Mattea, F. and Varona, S. (2009) Encapsulation and co-precipitation processes with supercritical fluids: fundamentals and applications. *Journal of Supercritical Fluids*, **47**, 546–555.

Gomes, M.T.M.S., Santos, D.T. and Meireles, M.A.A. (2012) Trends in particle formation of bioactive compounds using supercritical fluids and nanoemulsions. *Food and Public Health*, **2**, 142–152.

Herrero, M., Plaza, M., Cifuentes, A. and Ibáñez, E. (2010) Green processes for the extraction of bioactives from rosemary: chemical and functional characterization via ultra-performance liquid chromatography-tandem mass spectrometry and *in-vitro* assays. *Journal of Chromatography A*, **217**, 2512–2520.

Huang, E.T.S., Cape, S.P., Alargov, D.K., Villa, J.A., Meresman, H., Rinner, L., Liang, C. and Sievers, R.E. (2003) Nanoparticle and microparticle generation with super or near critical carbon dioxide. In: 6th International Symposium on *Supercritical Fluids*, Versailles, France.

Ibáñez, I., Cifuentes, A., Rodriguez-Meizoso, I., Mendiola, J.A., Reglero, G., Señorans, F.J. and Turner, C. (2009) Device and process for the on-line extraction and drying of complex extracts. Spanish Patent P200900164.

Martín, Á. and Weidner, E. (2010). PGSS-drying: mechanisms and modeling. *Journal of Supercritical Fluids*, **55**, 271–281.

Martín, Á., Varona, S., Navarrete, A. and Cocero, M.J. (2010a) Encapsulation and co-precipitation processes with supercritical fluids: applications with essential oils. *The Open Chemical Engineering Journal*, **4**, 31–41.

Martín, Á., Pham, H.M., Kilzer, A., Kareth, S. and Weidner, E. (2010b) Micronization of polyethylene glycol by PGSS (particles from gas saturated solutions)-drying of aqueous solutions. *Chemical Engineering and Processing*, **49**, 1259–1266.

Meterc, D., Petermann, M. and Weidner, E. (2008) Drying of aqueous green tea extracts using a supercritical fluid spray process. *Journal of Supercritical Fluids*, **45**, 253–259.

Nunes, A.V.M. and Duarte, C.M.M. (2011) Dense CO_2 as a solute, co-solute or co-solvent in particle formation processes: a review. *Materials*, **4**, 2017–2041.

Padrela, L., Rodrigues, M.A., Velaga, S.P., Fernandes, A.C., Matos, H.A. and Azevedo, E.G. (2010) Screening for pharmaceutical cocrystals using the supercritical fluid enhanced atomization process. *Journal of Supercritical Fluids*, **53**, 156–164.

Paz, E., Martín, A. and Cocero, M.J. (2012) Formulation of β-carotene with soybean lecithin by PGSS (particles from gas saturated Ssolutions)-drying. *Journal of Supercritical Fluids*, **72**, 125–133.

Perrut, M. (2004) Applications of supercritical fluid solvents in the pharmaceutical industry. In: Marinsky, J.A., Sen Gupta, A.K. and Marcus, Y. (eds), *Ion Exchange and Solvent Extraction. A Series of Advances*. CRC Press, Champigneulles, France, pp. 1–35.

Pham, M., Pollak, S. and Petermann, M. (2012) Micronisation of poly(ethylene oxide) solutions and separation of water by PGSS-drying. *Journal of Supercritical Fluids*, **64**, 19–24.

Prinn, K.B, Costantino, H.R. and Tracy, M. (2002) Statistical modeling of protein spray drying at the lab scale. *AAPS Pharmacological Science and Technology*, **3**, 1–8.

Reibe, C., Alessi, P., Knez, Z. and Weidner, E. (2008) Micronization of high viscous biopolymers. In: Proceedings of the 11th European Meeting on *Supercritical Fluids*, Barcelona, Spain.

Reverchon, E. (2002) Supercritical-assisted atomization to produce micro- and/or nanoparticles of controlled size and distribution. *Industrial and Engineering Chemistry Research*, **41**, 2405–2411.

Santos, D.T. and Meireles, M.A.A. (2010) Carotenoid pigments encapsulation: fundamentals, techniques and recent trends. *The Open Chemical Engineering Journal*, **4**, 42–50.

Sellers, S.P., Clark, G.S., Sievers, R.E. and Carpenter, J.F. (2001) Dry powders of stable protein formulations from aqueous solutions prepared using supercritical CO_2-assisted aerosolization. *Journal of Pharmaceutical Sciences*, **90**, 785–797.

Sievers, R.E., Best, J.A. and Cape, S.P. (2008) Human-powered dry powder inhaler and dry powder inhaler compositions. US Patent 0035143 A1.

Sievers, R.E., and Karst, U. (1997) Methods for fine particle formation. US Patent 5,639,441.

Sievers, R.E., Karst, U., Milewski, P.D., Sellers, S.P., Miles, B.A., Schaefer, J.D., Stoldt, C.R. and Xu, C.Y. (1999) Formation of aqueous small droplet aerosols assisted by supercritical carbon dioxide. *Aerosol Science and Technology*, **30**, 3–15.

Sievers, R.E., Milewski, P.D., Sellers, S.P., Miles, B.A., Korte, B.J., Kusek, K.D., Clark, G.S., Mioskowski, B. and Villa, J.A. (2000) Supercritical and near-critical carbon dioxide assisted low-temperature bubble drying. *Industrial and Engineering Chemistry Research*, **39**, 4831–4836.

Sievers, R.E., Huang, E.T.S., Villa, J.A., Kawamoto, J.K., Evans, M.M. and Brauer, P.R. (2001) Low-temperature manufacturing of fine pharmaceutical powders with supercritical fluid aerosolization in a bubble dryer. *Pure and Applied Chemistry*, **73**, 1299–1303.

Sievers, R.E., Huang, E.T.S., Villa, J.A., Engling, G. and Brauer, P.R. (2003) Micronization of water-soluble or alcohol-soluble pharmaceuticals and model compounds with a low temperature bubble dryer. *Journal of Supercritical Fluids*, **26**, 9–16.

Sievers, R.E., Quinn, B.P., Cape, S.P., Searles, J.A., Braun, C.S., Bhagwat, P., Rebits, L.G., McAdams, D.H., Burger, J.L., Best, J.A., Lindsay, L., Hernandez, M.T., Kisich, K.O., Iacovangelo, T., Kristensen, D. and Chen, D. (2007) Near-critical fluid micronization of stabilized vaccines, antibiotics and anti-virals. *Journal of Supercritical Fluids*, **42**, 385–391.

Skala, D. and Orlovic, A. (2006) Particle production using supercritical fluids. In: Hsu, J.-P. and Spasic, A.M. (eds), *Micro-, Nano-, and Atto-Engineering*. CRC Press, Belgrade, Serbia and Montenegro, pp. 641–678.

Tabernero, A., Valle, E.M.M. and Galán, M.A. (2012) Supercritical fluids for pharmaceutical particle engineering: methods, basic fundamentals and modelling. *Chemical Engineering and Processing*, **60**, 9–25.

Varona, S., Martín, A. and Cocero, M.J. (2011) Liposomal incorporation of lavandin essential oil by a thin-film hydration method and by particles from gas-saturated solutions. *Industrial and Engineering Chemistry Research*, **50**, 2088–2097.

Varona, S., Kareth, S., Martín, Á. and Cocero, M.J. (2010) Formulation of lavandin essential oil with biopolymers by PGSS for application as biocide in ecological agriculture. *Journal of Supercritical Fluids*, **54**, 369–377.

Weidner, E. (2009) High pressure micronization for food applications. *Journal of Supercritical Fluids*, **47**, 556–565.

Weidner, E., Kilzer, A., Petermann, M., Pross, A., Lucas, H.W. and Stepanski, H. (2000) Verfahren zur Erzeugung von Polyurethanpartikeln (Process for producing polyurethane particles). Deutsches Patent DE 100 40 551.7.

12

Flavouring and Coating Technologies for Preservation and Processing of Foods

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12.1 Introduction

The food industry, where food processing represents one of the most important stages, always seeks new technologies in order to ensure the quality and safety of food products. These are possibly the factors that most influence the decisions of consumers at the moment of buying a food product. Food technology plays a crucial role in seeking new know-how and the improvement of food quality during processing, storage and handling, while also maintaining its safety. It is known that conventional food sterilization and preservation methods often result in a number of undesired changes in foods, such as loss of odour, colour, flavour, texture and nutritional value. One of the ways to control and prevent food changes during processing and enhance food characteristics is the application of additives, which can be used with different objectives (e.g., preservation, colouring, improving nutritional value, texturizing and flavouring).

Flavour is one of the most important attributes in food products, which is often altered during processing, thus leading to the need to include flavouring

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additives in the formulation (Grandison, 2006). Flavours are among the most valuable ingredients added to food products; moreover, they can greatly influence, even in small quantities, the food products in several ways (e.g. quality and cost) (Madene *et al.*, 2006). They are some of the most added compounds to food products, not only to enhance their flavour but also to improve food preservation, since they can also be potent antioxidants, antimicrobials and nutraceuticals (Rijk, 2007; Pokorny and Trojáková, 2001).

Flavour science and food industry is continuously developing ingredients, processing methods and packaging materials to improve flavour preservation and delivery in order to increase flavour stability in foods, increasing their quality and acceptability. Most of the flavours are volatile and chemically unstable to air, light, moisture and high temperatures; moreover, they can be affected by interactions with food components (Gharsallaoui *et al.*, 2007; Madene *et al.*, 2006). Different flavour protection and application strategies have been industrially used; the most common is encapsulation of flavour compounds through a great number of methodologies (e.g. spray drying and extrusion). Coatings, commonly used to protect food from environmental aggressions and extending shelf-life, can also be used to encapsulate flavours and other functional additives in order to improve food properties during and after processing. This 'active packaging' solution has been developed to take advantage of the properties of the coating system like edibility, high compound retention and control release.

A flavour is a chemical compound or a mixture of several chemical compounds that has a smell or odour. They can be found in great numbers in food, wine, spice, perfume, fragrance oil and essential oil. Their application in the food industry is not trivial and their performance is usually the result of the presence of many volatile components of various chemical and physicochemical properties; moreover, food processing methods can have various sensorial impacts depending on the properties of each compound.

The stability of flavours and bioactive compounds in food products has been a matter of increasing interest. The manufacturing and storage processes, packaging materials and the type of ingredients used in foods can change their overall flavour and nutritional value. Despite flavour technology being well established, the encapsulation of new products, as well as the application of other methodologies, should be further developed or improved with the purpose of improving food flavouring performance. Edible coatings are one of the alternatives to replace conventional methodologies, since they can be used to improve appearance and quality of foods, as well as increase their safety.

In this chapter, after describing the main aspects of flavouring science, the utilization of edible coatings in foods and the application of flavours using coating technology are detailed. Finally, regulatory aspects in different countries and organizations are mentioned.

12.2 Flavouring of foods

Flavour is usually the result of the presence, within complex matrices, of volatile and nonvolatile components with a great range of physicochemical properties (Longo and Sanromán, 2010). Actually, flavours represent more than 25% of the world market for food additives and most of the flavouring compounds are produced via chemical synthesis or by extraction from natural materials. The main function of flavours is providing taste and/or smell to foods. The nonvolatile compounds contribute mainly to the taste while the volatile ones influence both taste and aroma (Longo and Sanromán, 2010; Abegaz *et al.*, 2004; Flores *et al.*, 1997).

12.2.1 Flavours used in preservation and processing of foods

Flavour additives are used for specific needs; examples include making a low flavour-impact food more appetizing, giving a specific attribute to a food that is produced from several component materials, restoring the integrity of flavours that have been adversely affected by processing conditions, making more appealing pharmaceuticals or nutraceuticals (e.g. vitamin C and multivitamin tablets that are coated with a sweet taste) (Omobuwajo, 2007), controlling food-borne pathogens and protecting food products (e.g. essential oils as food preservative agents) (Turgis *et al.*, 2012). Flavours should be compatible with other ingredients, able to resist processing conditions (e.g. extreme temperature and pressure, irradiation, vacuum and pH) while retaining their properties and be stable after processing. Furthermore, flavours must meet legal guidelines such as all relevant safety regulations. The acceptability by the consumer is also an extremely important point, since the whole concept of using flavourings in food is markedly influenced by consumer preference and acceptability. A flavour that meets most of the previous criteria will be more probably accepted by the consumer (Omobuwajo, 2007; Clark, 1998; Antoun and Tsimidou, 1997). The most important requirements for flavours used in foods are summarized in Figure 12.1.

Retention of added flavour characteristics will be influenced by processing, packaging, storage, distribution and merchandising conditions; therefore, it is extremely important to correctly select the form used and the point of introduction of flavourings in the process.

In some cases, the order of succession of such operations cannot be altered without changing the materials added. In situations where the rearrangement of operations is not possible, flavourings must be added close to the end of the processing operation in order to limit exposure to damaging conditions. Whenever rearrangement is possible, flavourings should be mixed with the other components in order to minimize damaging them during processing.

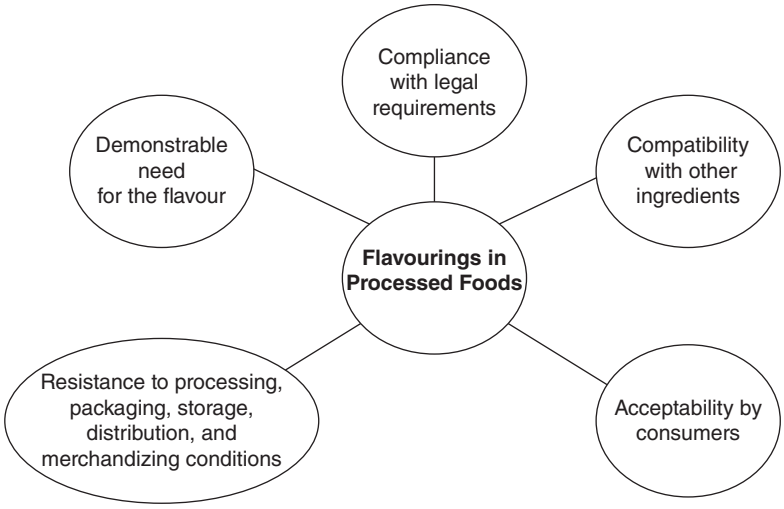

Figure 12.1 Main requirements for flavours used in foods

Usually most flavourings can be obtained in different forms: as liquids (Madene *et al.*, 2006), as powders (Bonvehí, 2005) and encapsulated (de Roos, 2003).

The deterioration of food quality results from a wide range of reactions, which can be physical, chemical, enzymatic and microbiological. It is possible to avoid food spoilage and eventual poisoning caused by those reactions using a number of preservation techniques, most of which act by preventing or slowing down reaction rates/microbial growth (Cleveland *et al.*, 2001). Food preservation can be achieved by promoting a longer shelf-life using techniques such as freezing, chilling, drying, curing, vacuum packing, modified atmosphere packaging, acidifying, fermenting, or adding chemical or natural preservatives (e.g. plant-derived antimicrobials) (Neetoo and Chen, 2012; Gould, 2000). In the last decade, the food industry has focused on procedures that deliver food providing a high level of microbial safety, good organoleptical quality and nutritionally healthy, while minimizing the use of chemical preservatives (Cleveland *et al.*, 2001; Dorman and Deans, 2000). For example, spices and herbs, which are currently used as flavouring and seasoning agents in foods, not only help preserving food due to their natural antimicrobial and antioxidant properties but also add flavour (Gupta and Nair, 2012). In this situation, the constituents with antimicrobial and/or antioxidant properties are found in different fractions; some of them are present in the volatile fraction (e.g., carvacrol, timol and eugenol) and others are in the non-volatile fraction (e.g., curcumin, capsaicine and carnosic acid) (Regnier, Combrinck and Du Plooy, 2012; Shinde and Nagarsenker, 2011; Lakkis, 2007).

Volatile essential oils (EOs) have great potential as food antimicrobials. These are mostly obtained from spices and herbs, and are mainly responsible for flavouring in foods (Regnier, Combrinck and Du Plooy, 2012; Juneja, Dwevedi and Yan, 2012; Hamad, 2012; Albarracin, Alfonso and Sanchez, 2012). Additionally, scientific evidence suggests that these oils possess strong antioxidant activities, which are favourable properties to combat free radical-mediated organoleptic deterioration; when added to food, they would retard microbial contamination and therefore reduce the onset of spoilage (Gupta and Nair, 2012; Yanishlieva, Marinova and Pokorný, 2006; Cutter, 2000). Approximately 3000 EOs, of which 300 are commercially available, are currently documented (Hyldgaard, Mygind and Meyer, 2012; Bakkali *et al.*, 2008; Burt, 2004).

Despite the minimum inhibitory concentration (MIC) levels of some EOs when used in food products, they need to be two- to tenfold higher than the concentrations tested *in vivo* in order to achieve the same antibacterial effect (Burt, 2004).

Moreover, the gram-positive bacteria seem to be more susceptible to EOs when compared with gram-negative bacteria (Inouye, Takizawa and Yamaguchi, 2001; Tassou and Nychas, 1995). Despite all the EOs advantages, they also exhibit concerns associated with their use; for example, EOs exhibit an intense odour at unacceptable levels and inappropriate flavours when used at effective doses (Belletti *et al.*, 2010; Tassou, Koutsoumanis and Nychas, 2000). Some of the studied flavours that showed antibacterial and antifungal activities are presented in Table 12.1.

Once again, the nonvolatile fraction of the flavours obtained from plants and other products is the main part used in food preservation. Some examples of flavouring ingredients that can be used as preservatives due to their antioxidant, antimicrobial or other biological activities are coffee, tea, cocoa and coca leaves (Attokaran, 2011; Soher *et al.*, 2011). Moreover, nonvolatile secondary metabolites contain some of the most important bioactive compounds, such as amino acids, lectins, glycoproteins, flavonoids, tannins, quinones, coumarins, terpenoids, steroids and alkaloids (Regnier, Combrinck and Du Plooy, 2012; El-Ghorab, El-Massry and Shibamoto, 2007). For example, nonvolatile fractions of EOs (e.g. extracted from orange) may possess antioxidant activities that could be used for stabilization of the volatile fractions (Pokorny and Trojáková, 2001). It has been reported that EO of orange has 1% of nonvolatile compounds, such as carotenoids, tocopherols, flavonoids, hydrocarbons, fatty acids and sterols (Cabral *et al.*, 2010). Onion, due to its high content of flavonoids, may be useful to improve food preservation without undesirable changes of organoleptic properties (Santas, Almajano and Carbó, 2010). Nonvolatile compounds commonly found in small fruits may include ascorbic acid (vitamin C), citric acid, malic acid, tartaric acid, and calcium and potassium salts (Perkins-Veazie and Collins, 2001).

Table 12.1 Essential oils (EOs) and respective antimicrobial activity as potential food preservatives

Plant from which EO is derived	Microorganisms	Activity	Reference
Callistemon lanceolatus	*Aspergillus flavus*	Antifungal	Shukla *et al.* (2012)
Onion	*Escherichia coli*, *Bacillus subtilis*, *Saccharomyces cerevisiae*, *Aspergillus niger*	Antibacterial, antifungal	Ye, Dai and Hu (2013)
Clove	*E. coli*, *B. subtilis*, *A. flavus*, *Mucor* sp.	Antibacterial, antifungal	Pundir, Pranay and Chetan (2010)
Garlic	*B. subtilis*, *Staphyloccus aureus*, *E. coli A. flavus*, *Mucor* sp.	Antibacterial, antifungal	Pundir, Pranay and Chetan (2010)
Cinnamon	*A. flavus*	Antifungal	Tian *et al.* (2012)
Coriander, common myrrh, *Cananga odorata*	*A. flavus*	Antifungal, antibacterial	Prakash *et al.* (2012)
Mount Atlas mastic	*Salmonella typhimurium*, *E. coli*, *Staphylococus epidermidis*, *B. subtilis*	Antibacterial	Mohagheghzadeh, Faridi and Ghasemi (2010)
Mint Thyme	*Micrococcus flavus*, *S. typhimurium*	Antibacterial	Soković *et al.* (2010)
Zizyphus jujuba	*S. aureus, B. subtilis*, *E. coli*	Antibacterial	Al-Reza *et al.* (2010)
Artemisia anomala S.	*E. coli*, *S. typhimurium*, *B. subtilis*	Antimicrobial	Guangrong, Jiaxin and Dehui (2008)
Ginger oil	*B. subtilis*, *Pseudomonas aeruginosa*, *A. niger*	Antifungal, antibacterial	Sasidharan and Menon (2010)

Oleoresins (a resinous–viscous natural concentrated liquid of flavourings) contain both volatile and nonvolatile flavour components (Lee and Lee, 2003). Spices contain many oleoresins, which are particularly useful since they encompass many of the non-volatile components that are not present in the corresponding essential oils (Knights, 2010). The highest level of attention among herbs and spices as sources of antioxidant activity has been focused on rosemary (*Rosmarinus officinalis* L.). Rosemary plants have many phytochemicals, which constitute potential sources of natural compounds (e.g. phenolic diterpenes, flavonoids and phenolic acids) (Moreno *et al.*, 2012; Erkan, Ayranci and Ayranci, 2008; Pokorny and Trojáková, 2001). Sage (*Salvia officinalis* L.) has been shown to have similar patterns of phenolic compounds and similar antioxidant activity to rosemary (Erkan, Ayranci and Ayranci, 2008). In oregano herbs oleoresin contains many valuable non-volatile constituents like urosolic acid, oleanolic acid, protocatechnic acid,

Table 12.2 Sources and applications of nonvolatile compounds

Food products	Examples	Notes	References
Grapes	Sweet basil, fennel, summer savory and thyme	Good antifungal effect, reduction of weight loss and have no effect on the flavour	Abdollahi *et al.* (2012)
Tomato puree	Garlic and clove	Increase the shelf-life for a maximum of ten days	Martin-Sánchez *et al.* (2011)
Sausages	Oregano	No effect on the sensory properties, improve the texture	Martin-Sánchez *et al.* (2011)
Pork loin and belly	Garlic and onion	Inhibit the growth of bacteria and the production of oxidative products	Park *et al.* (2008)
Cod fillets	Oregano and cinnamon	Reduction of *Photobacterium phosphoreum* growth in cod fillets and extension of shelf-life at 2°C	Mejlholm and Dalgaard (2002)
Nile tilapia fillet	Rosemary and thyme	High effectiveness as antioxidants, even at low concentrations	Albarracín, Alfonso and Sanchez (2012)
Yogurt drink	Mint	Effective for inactivation of *Listeria monocytogenes* and *L. innocua* without any adverse effect on pH	Evrendilek and Balasubramaniam (2011)

tilianin, sagittatoside, daucosterol, β-sistosterol and stigmasterol (Attokaran, 2011). Nonvolatile phytochemical compounds of thyme can be used as natural preservative ingredients in the food industries (Sarikurkcu *et al.*, 2010). Other sources and applications of nonvolatile compounds in fruit, meat, fish and dairy products are shown in Table 12.2. Both volatile and nonvolatile flavour components are responsible for influencing the aromatic descriptors on food products so that their characteristics and interactions play an important role in determining the overall flavour (Taylor, 1998).

12.2.2 Methodologies for flavour encapsulation

Encapsulation has been used in the food industry for more than 60 years and has received increased interest with the evolution from micro- to nano-encapsulation (Shimoni, 2009). This technology changes its denomination according to the final capsule size. Therefore, the product obtained by this process is called a macrocapsule when the particle size is larger than 5000 μm, a microcapsule when the size ranges between 1.0 and 5000 μm

and a nanocapsule when the size is below 1 μm (Jafari *et al.*, 2008). The main interest in reducing the size of the capsules was to limit the impact of capsule addition on physical and organoleptic properties as well as improving delivery and release performances of the active agents.

Food matrix components can also entrap or encapsulate volatile or non-volatile flavour compounds. The affinity of the flavour compounds with the food matrix is extremely important because it will affect the flavour delivery process. As a result of encapsulation, the rate of flavour release is reduced and it is possible to control flavour intensity and quality of foods (Naknean and Meenume, 2010). The encapsulation process should be done prior to use in foods or beverages in order to protect food flavourings limiting aroma degradation or loss during processing and storage. Moreover, it will influence the overall acceptance by consumers (Naknean and Meenume, 2010; Madene *et al.*, 2006).

Encapsulation is responsible for creating a befitting microenvironment around flavours and has several advantages (Figure 12.2) (Kralovec *et al.*, 2012; Fang and Bhandari, 2012; Xie, Zhou and Zhang, 2007; Gharsallaoui *et al.*, 2007; Madene *et al.*, 2006; Gouin, 2004; Che Man, Urwandi and Abdulla, 1999; Druaux and Voilley, 1997). It can also prevent off-tastes and mask strong or bitter flavours (e.g. green tea is sometimes encapsulated to mask the off-flavour that may develop with oxygen reacting with fatty acids) (Onwulata, 2012; Nedovic *et al.*, 2011).

After a general encapsulation process, the coated or entrapped compound is called active or core material, and the 'exterior' coating material is called

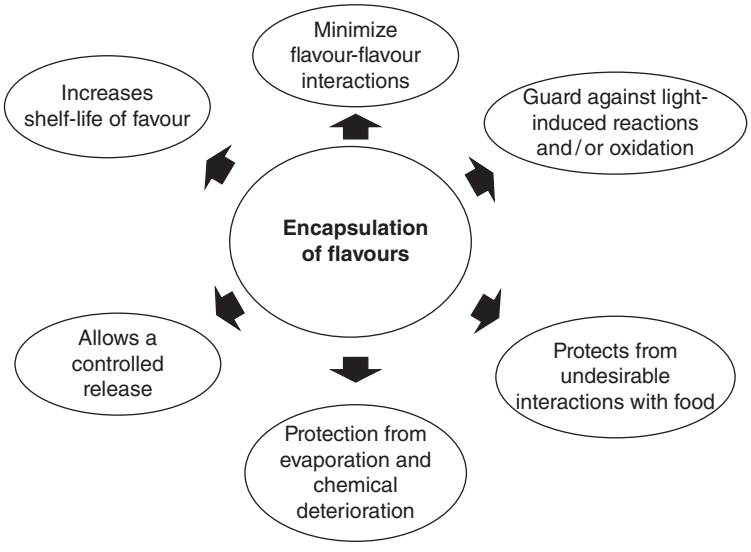

Figure 12.2 Advantages of flavour encapsulation

wall material, shell, carrier or encapsulant (Fang and Bhandari, 2012; Madene *et al.*, 2006). The process for encapsulation of sensitive compounds consists of two steps: the first is often emulsification of a core material, such as the 'lipid-aroma' system, with a dense solution of a wall material such as a polysaccharide or protein. The second is drying or cooling of the emulsions (Madene *et al.*, 2006). Encapsulation techniques can be divided into three classes: (1) physical processes such as spray drying, spray chilling/coating and extrusion; (2) chemical processes such as molecular inclusion or interfacial polymerization; and (3) physicochemical techniques such as coacervation and liposome encapsulation (Onwulata, 2012; Kralovec *et al.*, 2012; Desai and Park, 2005a). These techniques have been applied with ever-increasing popularity (Yeo *et al.*, 2005); however, the two major industrial processes used in flavour encapsulation are spray drying and extrusion (Nedovic *et al.*, 2011; Manojlovic *et al.*, 2008).

Matrices are considered an important condition to preserve the properties of the flavour materials and the encapsulation process employed. They will have different shapes (e.g. films, spheres and irregular particles), structures (porous or compact) and different physical structures (amorphous or crystalline dehydrated solid, rubbery or glassy matrix). All these previous conditions will influence diffusion of flavours or external substances (oxygen and solvent) as well as the food product's stability during storage (Madene *et al.*, 2006; Shaikh, Bhosale and Singhal, 2006).

To ensure a successful encapsulation of flavour compounds the carrier material should respect several conditions, as indicated in Figure 12.3 (Madene *et al.*, 2006).

In the case of wall or encapsulant material, there are several options such as maltodextrin, hydrophobically modified starch, gum Arabic (Fang and Bhandari, 2012; Abbas *et al.*, 2012; Shaikh, Bhosale and Singhal, 2006) and mixtures (Abbas *et al.*, 2012; Fang and Bhandari, 2012), such as globular proteins with anionic polysaccharides (Kralovec *et al.*, 2012; Weinbreck *et al.*, 2004). Also used are alginate, gelatine, gliadin, carrageenan, polyvinyl alcohol (Fang and

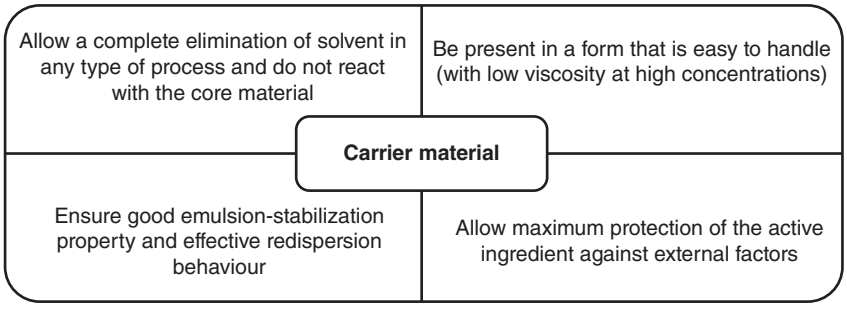

Figure 12.3 Characteristics of carrier material in the encapsulation process

Bhandari, 2012; Thies, 2007), modified cellulose, cyclodextrin, lecithin and hydrogenated fat, among others; the utilization of whey proteins may also be considered since they exhibit excellent microencapsulating properties and are suitable for microencapsulation of volatile and nonvolatile core materials (Baranauskiene *et al.*, 2007; Madene *et al.*, 2006; Beristain, García and Vernon-Carter, 2001). Comparatively low water solubility polysaccharides, Maillard reaction products and esters of fatty acids can also be used as encapsulant materials (Abbas *et al.*, 2012).

An area of increasing research interest is the development of alternative and inexpensive natural materials. These can encapsulate flavours with higher or at least the same efficiency, although their development as encapsulating agents is still challenging (Abbas *et al.*, 2012). There is a need to search for other natural gums with a focus on their emulsification and shell properties. Mesquite gum, for instance, has been shown to impart a better stability of oil/water (o/w) emulsions and higher encapsulation efficiency when compared to gum Arabic (Beristain, García and Vernon-Carter, 2001, 1999). It was also reported that the use of depolymerized guar gum as wall material is possible; guar gum was successfully used as a partial substitute of gum Arabic for microencapsulation of mint oil (Sarkar *et al.*, 2012).

Spray drying Encapsulation using spray drying is widely used by food industries. More than 90% of the flavours available in the market are produced or encapsulated by spray drying (Murugesan and Orsat, 2012). This technique has been used since the 1950s to convert liquids to powders and also to provide flavour oils with some protection against degradation/oxidation (Desai and Park, 2005a). Among the different encapsulation techniques, spray drying is extensively used for microencapsulation of food ingredients and food preservation; besides being flexible, simple, continuous and fast, it is also an economical operation (Beirão da Costa *et al.*, 2012; Murugesan and Orsat, 2012; Fang and Bhandari, 2012; Kralovec *et al.*, 2012; Onwulata, 2012; Nedovic *et al.*, 2011; de Vos *et al.*, 2010; Xie *et al.*, 2007; Shaikh, Bosale and Singhal, 2006; Desai and Park, 2005). It is broadly employed in large-scale production of encapsulated flavours and volatiles (e.g. encapsulation of flavour oils) (Fang and Bhandari, 2012; Krishnan, Kshirsagar and Singhal, 2005).

The process involves forming a matrix layer with the encapsulating material and forcing core materials into the matrix using a spinning atomizer; the final result is the formation of multicomponent spheres (Onwulata, 2012). The flavour compounds are encapsulated within the core materials in the drying medium. The hot or cold medium evaporates the dried powder containing the encapsulate within the core material. Then the resulting microcapsules are collected after they fall to the bottom of the drier (Onwulata, 2012; Desai and Jin Park, 2005). Typically, the final spray-dried particles have a mean size range of 10–100 μm (Fang and Bhandari, 2012). Likewise, the selection of appropriate coating materials, the drying process and preparation of emulsions with core

Table 12.3 Advantages and disadvantages of microencapsulation techniques

Technique	Advantages	Disadvantages
Spray drying	Relative gentle methodology in terms of solvents and matrix molecule applications	Limited availability and high cost of the wall or encapsulating materials
	Usually the product is very stable and allows a significant increase in shelf-life	The oil loading level is low and air inclusion in the emulsification process is difficult
	Friendly technique for food manufacturers	Requires high temperatures during the immobilization process
Coacervation	Simple and solvent-free process; highly advantageous for the production of nanoparticles in industry	Very expensive and rather complex process
	High payloads (up to 99%) and controlled release possibilities	Crosslinking of the wall material usually involves glutaraldehyde, which must be carefully used according to a country's legislation
Extrusion	Encapsulation is relatively gentle, and does not involve harmful solvents	Limited to the high boiling point of flavours
	Stability of the flavour against oxidation and therefore prolongs the shelf-life	To expand the application to low boiling point flavours, isopropanol is added to the process, which can be hazardous
	True encapsulation procedure instead of an immobilization technology	The microcapsule may contain microporosity when low boiling point components are present in the flavour

and coating materials influence the shape and type of capsule formed, thus affecting the rate of retention of core compounds (Onwulata, 2012).

Overall, spray drying has advantages over other technologies for encapsulating bioactive food components (e.g. as flavours) (de Vos *et al.*, 2010; Che Man, Irwandi and Abdulla, 1999); however, it also has some limitations (Murugesan and Orsat, 2012; Kralovec *et al.*, 2012; de Vos *et al.*, 2010; Drusch *et al.*, 2007), which are presented in Table 12.3.

Coacervation This technique can be classified as simple coacervation if it only involves one type of polymer; when two or more types of polymers of opposite ionic charges are present, it is named *complex coacervation* (Fang and Bhandari, 2012; Prata *et al.*, 2008).

Coacervation encapsulation may be stated as the phase separation of one or many hydrocolloids from the initial sample dispersion and the subsequent deposition of the newly formed coacervate phase around the active ingredient suspended/emulsified in the same media (Fang and Bhandari, 2012; Gouin,

2004). Complex coacervation balances the electrostatic interaction between the two components of the encapsulation emulsion in order to create water- and heat-resistant capsules; a typical complex coacervation process begins with the dispersion or emulsification of core material (e.g. in either gelatin or gum Arabic dispersion). Subsequently, when the core material dispersion is mixed with an oppositely charged encapsulating material, a complex is formed, resulting in phase segregation and associative complexation (Onwulata, 2012). Capsules result from the ability of the coacervate to form a coating around sensitive materials. When coacervate droplets are formed, they usually coalesce and sediment as a separate (coacervate) phase. If an insoluble material (such as an oil droplet containing flavours) is present in the mixture, the coacervates will deposit at the surface of this material and, if sufficient stirring is applied to prevent sedimentation of the coacervate droplets, the compound to be encapsulated will be homogeneously coated by a layer of coacervate. A prerequisite is that the coacervate phase wets the particles or oil droplets (Gouin, 2004; de Roos, 2003). The size and other characteristics of the capsules formed can be controlled by changing pH, temperature, bioactive component properties and type of encapsulating agent. This technique is usually used to encapsulate water-insoluble liquids such as oil and flavourings (Onwulata, 2012; Fang and Bhandari, 2012; de Vos *et al.*, 2010). During coacervation of flavour compounds some parameters, such as the concentration of the polyanion solutions and the rate of homogenization, affect the morphology and size distribution of the microcapsules: (a) the rate of homogenization affects the size of oil cores encapsulated within the microcapsules whereas (b) the polyanion concentrations affect the number of core aggregates consisting of a microcapsule. Among all the complex coacervation systems, the most studied and well understood is the gelatine/gum Arabic system (Fang and Bhandari, 2012).

Recently, researches have been conducted to support the high potential of complex coacervation for designing functional microencapsulation systems. The challenge now is to increase the knowledge of complex coacervation methods and to develop standardized protocols for improving the quality of encapsulated products (e.g. flavours). New approaches for controlling the pH and temperature of coacervation systems would be beneficial. The adoption of a freezing process to control the kinetics of complex coacervation, especially when dealing with food ingredients appears pragmatic. An interesting example that has been recently developed is a room-temperature process for the encapsulation of heat-sensitive ingredients such as volatile flavour oils (Nakagawa and Nagao, 2012; Fang and Bhandari, 2012; de Vos *et al.*, 2010; Desai and Park, 2005a). Advantages and disadvantages associated with this technique are reviewed in Table 12.3.

Extrusion Another encapsulation technique is the category of extrusion technologies (de Vos *et al.*, 2010). Encapsulation of food ingredients by extrusion has been essentially used for the encapsulation of volatile and unstable flavours in glassy carbohydrate matrices. Hence, when compared with spray drying, extrusion is considered a relatively new process (Fang and Bhandari, 2012).

Extrusion methods consist of dropping droplets of an aqueous dispersion of polymer and active compound into a gelling bath. The dripping tool (or droplet-generating device) can be simply a pipette, a syringe, a vibrating nozzle, a spraying nozzle, jet cutter or atomizing disc; the smaller the inner diameter of the nozzle or openings, the smaller are the capsules produced (de Vos *et al.*, 2010). Extrusion was found to be an appropriate technology for large-scale/industrial applications. Moreover, large-scale droplet production can be achieved by multiple-nozzle systems, a spinning disc atomizer or by jet-cutter techniques (Nedovic *et al.*, 2011; de Vos *et al.*, 2010). It is a relatively low-temperature entrapping method that involves forcing a core material in a molten carbohydrate mass through a series of dies into a bath of dehydrating liquid. The pressure and temperature employed are typically less than 7 atmosphere and seldom above 115 °C. The coating material hardens on contacting the liquids, forming an encapsulating matrix to entrap the core material. Then the extruded filaments are separated from the liquid bath, dried and sized. The carrier used may be composed of more than one ingredient, such as sucrose, maltodextrin, glucose syrup, glycerine and glucose (Fang and Bhandari, 2012; de Vos *et al.*, 2010).

Extrusion has been successfully applied to flavour encapsulation (Fang and Bhandari, 2012; Byun *et al.*, 2010; Risch and Reineccius, 1988). Carbohydrate matrices have very good barrier properties and extrusion is a convenient process enabling the encapsulation of flavours (Fang and Bhandari, 2012; Manojlovic *et al.*, 2008). Microcapsules from 200 to 2000 μm have been produced by various extrusion techniques with a maximal flavour load going up to 20%, w/w (Manojlovic *et al.*, 2008; Madene *et al.*, 2006). Advantages and disadvantages of the extrusion technique are also shown in Table 12.3.

12.2.3 Applications

There are several techniques employed in flavour encapsulation. Table 12.4 summarizes applications published in the literature.

The main concern in the food flavour industry is processing cost. Hence, from an industrial point of view, spray drying is recommended as the preferred technique for the microencapsulation of food flavours. It is considered the most economical and flexible method for encapsulation. There is a need

Table 12.4 Applications for encapsulation of food flavours

Technique	Application	Conditions	Reference
Spray drying	Cardamom EO using Mesquite gum	Gum:oil (4:1); inlet air at 200 °C and outlet air at 110 °C; encapsulation efficiency of 83.6%	Beristain, García and Vernon-Carter (2001)
	Ginger oil powder using Acacia gum	Inlet air at 160 °C and outlet air at 108 °C; encapsulation efficiency of 91%	Kadam, Hashmi and Kale (2011)
	Vitamin C using tripolyphosphate crosslinked chitosan microspheres	Inlet air at 175 °C and outlet air at 108 °C; encapsulation efficiency ranged between 45.05 and 58.30%	Desai and Park (2005b)
Extrusion	Thyme aqueous extract using calcium alginate beads	Positively charged (6.5 kV) blunt stainless steel needle (22 gauge) at 25.2 mL/h, by a syringe pump; encapsulation efficiency ranged between 50 and 80%	Stojanovic *et al.* (2011)
	Polyphenolic antioxidants using alginate–chitosan	Positively charged (7.3 kV) blunt tip metal needle (23 gauge) at 25.2 mL/h, by a syringe pump; encapsulation efficiency ranged between 80 and 89%	Belščak-Cvitanović *et al.* (2011)
Coacervation	Bake flavour oil using gelatin and gum Arabic	Gelatin:gum Arabic (1:1); homogenization at 3000 and 9000 rpm; coacervation pH is 4	Yeo *et al.* (2005)
	Capsaicin using gelatin, acacia and tannins	Gelatin:Acacia (1:1) and 10% (w/v) aqueous dispersion of tannins; homogenization at 300 rpm; coacervation pH is 4.2; encapsulation efficiency of 88.2%	Xing *et al.* (2004)

for future research to focus on large-scale applications, improving existing technologies and also choosing new processing conditions and new carrier materials.

12.3 Edible coatings for food applications

Edible coatings enhance the quality of food products, protecting them from physical, chemical and biological deterioration (Rodriguez-Turienzo, Cobos and Diaz, 2012; Cerqueira *et al.*, 2009, 2010a; Kester and Fennema, 1986). Coatings can be defined as a film-forming solution that is directly applied on

to the surface, forming a thin film upon drying that will protect the food product (Han and Gennadios, 2005). The main differences between coatings and films is that a film is a stand-alone wrapping material, while a coating is a film-forming solution applied directly on the food surface itself (Pavlath and Orts, 2009).

12.3.1 Materials

The main components of everyday foods can be used as the main material for the production of edible coatings. Edible coatings are mainly produced from biopolymers and food-grade compounds (GRAS – generally recognized as safe) (Cerqueira *et al.*, 2011a; Han and Gennadios, 2005). The most used biopolymers are (Table 12.5) proteins (used to provide mechanical stability), polysaccharides (usually used to control oxygen and other gas transmissions (Martins *et al.*, 2012; Pereira *et al.*, 2010)), lipids and resins (even if they are

Table 12.5 Examples of polysaccharides, proteins and lipids used for edible coating formation

Type of material	Example	Reference
Polysaccharides	Alginate	Bierhalz, Altenholfen da Silva and Kieckbusch (2012)
	Chitosan	Cerqueira *et al.* (2012)
	Galactomannans	Cerqueira *et al.* (2012)
	Xanthan gum	Sun, Gunasekaran and Richards (2007)
	Pectin	Bierhalz, Altenholfen da Silva and Kieckbusch (2012)
	Carrageenan	Martins *et al.* (2012)
	Starch and modified starches	Cervera *et al.* (2004)
Proteins	Casein	Buonocore *et al.* (2003)
	Gelatin	Zhang *et al.* (2013)
	Collagen	Fadini *et al.* (2013)
	Corn zein	Ozcalik and Tihminlioglu (2012)
	Whey protein	Ramos *et al.* (2012)
	Soy protein	Jiang *et al.* (2012)
	Wheat gluten	Koehler, Kieffer and Wieser (2010)
Lipids/resins/waxes	Cocoa butter	Fadini *et al.* (2013)
	Bees wax	Yilmaz and Dagdemir (2012)
	Candelilla wax	Saucedo-Pompa *et al.* (2009)
	Carnauba wax	Chiumarelli and Hubinger (2012)
	Fatty acids	Zahedi, Ghanbarzadeh and Sedaghat (2010)
	Shellac resin	Soradech *et al.* (2012)
	Acetylated monoglycerides	Anker *et al.* (2002)

not considered to be biopolymers (Fadini *et al.*, 2013; Cerqueira *et al.*, 2012) they are usually used to reduce water transmission (Pavlath and Orts, 2009)). These materials can be used alone or mixed as a composite blend (Martins *et al.*, 2012; Cerqueira *et al.*, 2012; Lima *et al.*, 2010; Baldwin *et al.*, 1997). Their chemical structures can differ widely and therefore attributes of each component contribute to overall coating properties (Martins *et al.*, 2012; Soradech *et al.*, 2012).

To form a matrix with sufficient cohesion and continuity, the formulation of an edible coating implies using at least one material able to do it (Nussinovitch, 2003), ensuring that the produced coating presents the desired physical and chemical properties (Pavlath and Orts, 2009). Hydrogen bonds may play significant roles in film formation, thus influencing their characteristics, and are related to the large numbers of hydroxyl groups in the biopolymer structure. Starch, a common agricultural raw material, is the most used polysaccharide for biodegradable edible coating formulations, due to its appropriate matrix-forming properties, lower cost when compared with other alternatives, availability and relative ease of manipulation (Garcia *et al.*, 2012; Guilbert and Gontard, 2005). Protein film-forming materials are derived from animal tissues, milk, eggs, grains and oil seeds (Vicente *et al.*, 2011; Krochta, 2002, 1997). Lipids and resins are edible, biodegradable and cohesive water barrier biomaterials despite not being considered as biopolymers. Due to their hydrophobicity, coatings and films made from lipids have a low surface energy and very high water resistance (Zaritzky, 2011). They are commonly applied in thin layers or as composites with a polymeric matrix. In order to increase the resistance to water penetration, other materials such as proteins or polysaccharides can be combined, as emulsion particles or as multilayer coatings (Cerqueira *et al.*, 2012; Pérez-Gago and Krochta, 2005, 2000; Wu *et al.*, 2002). Table 12.5 presents examples of the most used materials for edible coating formations.

Films containing polysaccharides and proteins often have a brittle and stiff structure owing to extensive interactions between the polymer molecules (Krochta, 2002). The addition of plasticizers improves flexibility, extensibility and processability of the films and avoids their brittleness (Guilbert and Gontard, 1995). Plasticizers incorporated into the polymeric film-forming material are low molecular weight agents. Most of them are very hydrophilic and hygroscopic (Zaritzky, 2011). Plasticizers must be compatible with the film-forming polymer; they reduce the intermolecular forces and increase the mobility of polymer chains and the free volume of polymer structures (Sothornvit and Krochta, 2001, 2000) by decreasing the ratio between the crystalline and the amorphous regions and the glass transition temperature (T_g) of the polymer (Cerqueira *et al.*, 2012; Guilbert and Gontard, 2005; Krochta, 2002). Some common plasticizers used are glycerol, sorbitol,

sucrose, propylene glycol and polyethylene glycol (Cerqueira *et al.*, 2012; Zhang and Han, 2006; Bai *et al.*, 2003).

Better uniformity can be promoted by adding surfactants to solution in order to reduce surface tension. This strategy will also reduce the superficial water activity (a_w) and in turn reduce water loss (Han and Gennadios, 2005; Krotcha, 2002). Surfactants like Tween and Spans have been used and are widely described (Ramos *et al.*, 2012; Han and Gennadios, 2005; Krotcha, 2002). To maintain edibility, solvents used should be restricted to water and ethanol, and all coating-forming components should be food-grade (Zaritzky, 2011).

Recently, the utilization of edible coatings as a way to incorporate functional additives became one of the most studied technologies in packaging science. Active and intelligent packaging using edible coatings has several advantages when compared with synthetic and conventional packaging. Added functional additives may include flavours, antimicrobials, antioxidants, nutrients, nutraceuticals or colourants, which, when combined with film-forming biopolymers, may lead to structural modifications or new functionalities of coatings (Martins, Cerqueira and Vicente, 2012; Cerqueira *et al.*, 2011a; Martín-Belloso, Rojas-Graü and Soliva-Fortuny, 2009).

Bioactive compounds include alkaloid, anthocyanin, carotenoid, flavonoid, glucosinolate, isoflavone, phenolic acid, tannin and terpene phytochemicals, protein hydrolysates, green and black tea extracts and essential oils. Natamycin, nisin, α-tocopherol, tea extracts, essential oils, seed extracts, acetic acid, propionic acid, potassium sorbate and thymol are some of the additives that were used and tested (Ruiz-Navajas *et al.*, 2013; Martins, Cerqueira and Vicente, 2012; Martins *et al.*, 2012, 2010; Bierhalz, Altenhofen da Silva and Kieckbusch, 2012; Sun-Waterhouse and Wadhwa, 2012; Vodnar, 2012; Gniewosz and Synowiec, 2011; González *et al.*, 2011; Sayanjali, Ghanbarzadeh and Ghiassifar, 2011; Cerqueira *et al.*, 2010b; Ouattara *et al.*, 2000).

12.3.2 Properties

The challenge for the successful use of edible coatings is to stabilize their functional properties, which must be preserved throughout processing of a particular food, while their mechanical properties should be adapted according to the desired application. In order to be successful in the market, a coating must meet demands for legality, safety and performance.

The ideal materials for coating production must be chosen according to their mechanical, barrier and thermal properties, solubility and optical attributes. Therefore, coatings must be characterized and, depending of the measured property, it is often necessary to use coatings in the form of films.

Numerous techniques can be used to perform the characterization of the structure of biopolymers responsible for the formation of coatings. Examples are dynamic mechanical thermal analysis (DMTA), differential scanning calorimetry (DSC), X-ray diffraction and Fourier transform infrared spectroscopy (FTIR) (Zaritzky, 2011; Cerqueira *et al.*, 2011b).

Rheological analysis and viscoelastic properties of the dispersions help to determine the suitability of the food coating method. Rheological behaviours are normally estimated using viscometers and oscillatory rheometers that allow measurement of the viscoelastic parameters (Lopez, García and Zaritzky, 2008; García *et al.*, 2006, 2004). The most suitable method should be selected according to food characteristics, coating materials, objective of the coating and cost.

To achieve a satisfactory coating formulation factors like surface wetting and spreading are very important. Wettability depends on liquid intramolecular attractions and on the attraction of a liquid for a solid surface. It can be found by measuring the contact angle of the liquid with the solid surface; this measurement also indicates the surface hydrophobicity. The contact angle measurement is a simple technique for determining the relative difference between two surface tensions (Cerqueira *et al.*, 2009; Ribeiro *et al.*, 2007). Low-energy surfaces (high contact angles) are difficult to wet and can give poor results for a coating (Gilleo, 2007; Karbowiak, Debeaufort and Voilly, 2006). Cohesion is the attractive force between the molecules of a substance; it depends on the structure of the polymer used, its molecular length and weight distribution, geometry and on the position and type of the side groups, and influences the mechanical strength of films (Guilbert, Gontard and Gorris, 1996). Adhesion is also an important factor for the coating process; it is defined as the attractive force between the surface molecules of coating materials and food surfaces. Incomplete coating on the food surface can be caused by low adhesion. If the difference between the surface energy of the coating material and that of the uncoated surface is significantly large, it will lead to a weak coating process. The surface tension of the coating solution can be reduced by adding surface-active agents like emulsifiers (Ribeiro *et al.*, 2007; Guilbert and Gontard, 2005; Cuq, Gontard and Guilbert, 1995).

When a coating and/or film is formed it is very important to know its mechanical properties, such as tensile strength, elongation and elasticity, which can be measured by stretching a film to its breaking point, puncturing or measuring deformation using a texture measuring equipment (Cerqueira *et al.*, 2012; Pérez-Gago and Krochta, 2000; Park *et al.*, 1994). These properties are important because they allow the association of different advantages that each material exhibits and attain optimum properties. The tensile strength (TS) and the percentage of the elongation at break (EB) are influenced by the addition of surfactants, lipids, bioactive compounds and drying temperature changes (Bourbon *et al.*, 2012; Cerqueira *et al.*, 2012; Bravin, Peressini and Sensidoni, 2006; Phan *et al.*, 2002).

Barrier property of coatings is related to film structure and this is a critical parameter to improve the storage life of the products. Determination of the barrier properties of edible coatings should be characterized using stand-alone edible films determining transmission rates of specific migrants (McHugh and Krochta, 1994). Pertinent barrier properties of edible films include water vapour permeability (WVP) and oxygen and carbon dioxide permeability. Water vapour permeability is an important factor to consider when freshness of a food needs to be maintained during storage. Determination of WVP can be performed using commercial instruments; a large number of researchers applied the ASTM (1995) method E96 with the modifications introduced by Gennadios *et al.* (1994). Gas (CO_2 and O_2) barrier properties are a determinant for food coating applications because they affect the quality and physiological aspects of coated food during storage. O_2 permeability can be measured using commercial equipment. The gas permeability of films can be measured also by the accumulation method in a cell designed especially for that purpose (Bifani *et al.*, 2007; García *et al.*, 2000, 1999). This method was based on measuring the amount of gas diffusing through a film, quantified by gas chromatography.

Gas permeability is strongly related to crystallinity of polymeric chains; the higher the degree of crystallinity, the lower is the permeability as permeation occurs through the amorphous zones of the film. Thus, gas and vapour permeabilities depend not only on the ratio between crystalline and amorphous zones but also on chain mobility and specific interactions between the functional groups in the polymers and gases in the amorphous zones (Cerqueira *et al.*, 2012; Bourbon *et al.*, 2011; Miller and Krochta, 1997).

Water solubility is very important to determine possible applications for composite biopolymer coatings. It can be changed by adjusting the concentration of biopolymer in the film formulation in order to attend to the requirements of a specific application (Cerqueira *et al.*, 2012; Cuq *et al.*, 1996).

The evaluation of thermal properties through DSC, DMTA and TGA techniques can be used to determine the glass transition temperature (T_g), melting phenomena and the degradation behaviour of the materials (Cerqueira *et al.*, 2012; Martins *et al.*, 2012; Zaritzky, 2011). Glass transition is a reversible change that takes place in the polymer between the rubbery and glassy states, which are the properties of amorphous or semicrystalline materials modified when the temperature of the compounds rises above T_g. Generally, fully amorphous bioplastic applications are limited by the fact that T_g of a polymer is highly affected by the relative humidity (RH) (especially for hydrophilic polymers). Below T_g, the material is rigid and above T_g it becomes rubbery and viscoelastic (Mali *et al.*, 2006, 2005; Guilbert and Gontard, 2005; Roos and Karel, 1991).

Film opacity and colour parameters are a critical property of a food coating; such parameters significantly depend on the structure of the polymers used and their chemical composition and can influence the final applications of the

produced coating. Other techniques often used in order to relate chemical and physical properties, and microstructure of the coatings and films are X-ray diffraction, Fourier transform infrared spectroscopy, ultraviolet/visible (UV/Vis) spectroscopy and scanning electron microscopy (Cerqueira *et al.*, 2012; Martins, Cerqueira and Vicente, 2012; Souza *et al.*, 2009).

12.3.3 Application methods

Edible coating formulations must be wet when spread on the food surface; upon drying, the formulation must form a film coating with adequate adhesion, cohesion and durability to function properly (Martins *et al.*, 2010; Cerqueira *et al.*, 2009; Ribeiro *et al.*, 2007). The application and distribution of the film-coating material in a liquid form can be achieved using several methods like spraying, electrostatic, dipping, enrobing, paint brushing and pan coating (Khan *et al.*, 2012; Cerqueira, 2010; Guilbert, 1986). However, the selection of an appropriate method will depend on the materials used and on the intended effect of the coating.

For dipping applications, the product is directly dipped into the composite coating formulations, removed and allowed to dry, becoming a thin membranous film formed over the commodity. Pan coating involves a stainless steel pan that rotates in an inclined fashion. The coating is delivered by a pump to spray guns mounted in various parts of the pan. The coating is atomized by the spray guns (Grant and Burns, 1994).

When a thin and uniform coating is required for certain surfaces, spraying is useful (Cutter and Sumner, 2002). In fact, early coating procedures involved sprays, with further distribution over food surfaces via rollers or brushes, followed by tumbling to spread the coating evenly (Cutter and Sumner, 2002; Grant and Burns, 1994). In addition, spray coating may be used in combination with pan, fluidized bed and other coating techniques to deposit either thin or thick layers of aqueous solution or suspensions and molten lipids or chocolate. The spray nozzle plays a critical role in the coating process. The pressure, fluid viscosity, temperature and surface tension of the coating liquid and nozzle shape directly determine the spraying efficiency (Debeaufort and Voilley, 2009; Fellows, 2000). Electrostatic coating can also be used in order to achieve an effective coating application (e.g. higher transfer efficiency). This methodology is based on the charge of a powder using an electrical field that is then spread on the surface of a food, thus creating a well-distributed coating (Khan *et al.*, 2012; Amefia, Abu-Ali and Barringer, 2006). Some of the successful applications of this technique were reported on cheese and potato chips (Ratanatriwong, Barringer and Delwiche, 2003; Elayedath and Barringer, 2002)

12.3.4 Food applications

The structure, chemical composition and methods used to produce coatings are important and should be taken into account when selecting a coating to be applied in a specific food product. The storage conditions and properties of the food should also be considered (Zhao and McDaniel, 2005).

Several applications of coatings in food products are possible. These are fruit, vegetables (e.g. strawberries, apples, raspberries, tomatoes and carrots), meat, fish and cheese (Rodriguez-Turienzo, Cobos and Diaz, 2012; Mastromatteo, Conte and Del Nobile, 2012; Ramos-García *et al.*, 2012; García *et al.*, 2012; Qi *et al.*, 2011; Ustunol, 2009; Cerqueira *et al.*, 2009; Han *et al.*, 2004).

The overall objectives of these applications, depending on the food, are mainly delaying spoilage (Fajardo *et al.*, 2010), reducing water and moisture loss (Ramos *et al.*, 2012), preventing fungal growth (Saucedo-Pompa *et al.*, 2009), reducing oil absorption (Suarez *et al.*, 2008), reducing lipid oxidation (Rodriguez-Turienzo, Cobos and Diaz, 2012), reducing dehydration (Cerqueira *et al.*, 2010a), extending shelf- and storage-life (Qi *et al.*, 2011), reducing respiration activity (Lima *et al.*, 2010), delaying oxidative browning (Qi *et al.*, 2011), delaying colour changes and loss of firmness (Ramos *et al.*, 2012) and also reducing microbial contamination (Ramos-García *et al.*, 2012).

12.4 Food flavouring by coating

Edible coatings provide a physical barrier against mass transport from the environment to foods and from foods to the environment. These barrier properties are important for food protection. The consumers demand for a better food safety and higher nutritional and flavour properties. In recent years, active packaging has been developed to extend food shelf-life by improving and increasing coating properties and functionalities. For instance, more activity can be provided to edible coatings by adding active compounds, such as flavours. Flavours can be incorporated directly into the edible polymer matrix or can be encapsulated to protect their activity and properties.

12.4.1 Methodologies

In the past, a wide range of food products were technically unviable for manufacture but are now available due to significant efforts from both science and industry; one of the most prominent examples is encapsulation technology.

The initial step in encapsulating a food ingredient is the selection of a suitable coating material, basically a film-forming biopolymer that meets the safety requirements of governmental agencies such as the European Food

Safety Authority (EFSA) or the Food and Drug Administration (FDA) of the United States of America. In particular, for flavour and oil encapsulation the ideal coating material should meet several criteria, as mentioned above.

There are numerous techniques of encapsulation employed in the food industries as mentioned before; however, only three of them are used commercially for flavour encapsulation, that is spray drying, coacervation and extrusion. Among them, spray drying is the most extensively applied process in the food industry and, in particular, in encapsulation of flavours and oils.

The importance of imparting emulsifying properties depends upon the type of flavouring encapsulated, the encapsulation process and the final application of the encapsulated flavour. For instance, if flavouring is labelled as 'water soluble', the coating material must be a nonemulsifying compound such as maltodextrin. On the other hand, if flavours or any part thereof are insoluble in the encapsulation system used, then an emulsifying matrix is required. Emulsification is used to minimize flavour losses during the encapsulation process (i.e. spray drying and extrusion processes). There is a large amount of data in the literature showing that retention of water-insoluble flavourings are substantially improved if a good-quality emulsion is prepared and used during the encapsulation process (Hambleton *et al.*, 2008; Soottitantawat *et al.*, 2003; Risch and Reineccius, 1988). Good-quality emulsions are those with a mean particle size of about 1 μm. These emulsions are prepared using an emulsifying coating material such as acacia gum, a modified starch or protein. In terms of the final application, the majority of the flavour compounds used in the food industries is mainly in liquid form at room temperature. For food products and beverages such as cake and soup mixes, jelly crystals, dry beverage mixes and instant breakfast drinks, the use of liquid flavours is not technologically acceptable. Therefore, it is necessary to present the flavouring components in the form of a dry, free-flowing powder. Technologically speaking, encapsulation is widely used in dry flavour production since it can provide the convenience of a solid powder – for example, it is easily handled with reduced volatility and less oxidation (Jafari *et al.*, 2008). Examples of commonly used encapsulated flavours and oils are citrus oils, artificial or natural flavours, essential oils and spices, tuna oil, fatty acids, soy oil and sunflower oil (Frascareli *et al.*, 2012).

12.4.2 Influence of flavour incorporation on edible coating properties

Edible coatings can carry various active agents, such as flavourings, thus enhancing food quality and safety; however, flavouring incorporation can modify mechanical and barrier properties of edible coatings and films, because of physical changes induced in the network structure. Induced changes depend on factors such as the molecular size, polarity, shape, affinity

of the flavour incorporated and the polymer molecules of coatings and films (Han, 2002; Guilbert, Gontard and Gorris, 1996; Kester and Fennema, 1986).

Influence of flavouring agents on mechanical properties of edible coatings The incorporation of oil droplets into edible coatings usually induces a loss of mechanical properties, such as a decrease of the tensile strength (TS) and elastic modulus (EM). For instance, the addition of tea tree oil in a 0.5 to 3% concentration range causes a significant decrease in the EM and TS of hydroxypropyl methylcellulose films, although with no significant effect on elongation at break (EB) (Sánchez-González *et al.*, 2009). This is in agreement with the results reported by other authors when essential oils were incorporated into a chitosan matrix (Zivanovic, Chi and Draughon, 2005; Pranoto, Salokhe and Rakshit, 2005; Pranoto, Rakshit and Salokhe, 2005). Moreover, bergamot oil (0.5%) added to chitosan films reduced the TS and EB by twofold and threefold, respectively (Sánchez-González *et al.*, 2010a). On the other hand, cinnamon oil seemed to have some plasticizing effect on soy protein isolate films, since it makes them more extensible as the oil content increased (Atarés *et al.*, 2010). This effect was not observed when ginger oil was added to soy protein isolate films, which were less resistant and less elastic than those with cinnamon oil. A different behaviour was observed by Hambleton *et al.* (2012) for κ-carrageenan films when incorporated with *n*-hexanal, since the incorporation of this compound tends to increase the EM and TS. This is probably due to the stabilizing effect of this compound on the film matrix. *n*-Hexanal interacts with the lateral chains of κ-carrageenan and plays a stabilizing role in the interface due to its amphipolar character, leading to a much more homogeneous structure that increases the film stiffness. Nevertheless, *n*-hexanal does not affect the film capacity to stretch as the EB values are not significantly different from the same film without *n*-hexanal. On the contrary, incorporation of *n*-hexanal in sodium alginate films did not affect its mechanical properties because the flavouring agent weakly interacts with sodium alginate molecular chains (Hambleton *et al.*, 2009). This type of film has a well-organized structure, stabilized by divalent ions that form stronger gels and thus stronger films; however, *n*-hexanal may interact with other components of the film such as glycerol, which being a polyol has a great affinity for flavours of this type. These interactions lead to a reduction of the film stiffness and resistance to elongation. The presence of both *n*-hexanal and fat material has a significant effect, reducing the EM and TS more than in other types of films, probably because this aroma interacts primarily with the fat material when added to the film.

Influence of flavouring agents on the barrier properties of edible coatings The incorporation of flavourings or essential oils usually affects the oxygen and WVP properties of edible coatings and films. For instance,

when 1% *n*-hexanal was incorporated in carrageenan–glycerol and carrageenan–glycerol-lipid edible films, the oxygen permeability increased by 15 and 100%, respectively (Hambleton *et al.*, 2008). On the other hand, incorporation of 1% *n*-hexanal in sodium alginate films doubled the oxygen permeability and increased ten times when the film contained lipid emulsions (Hambleton *et al.*, 2009). Contrarily, Rojas-Graü *et al.* (2007b) did not show any change in the oxygen permeability of alginate–apple puree film when oregano, carvacrol, lemongrass oil, citral, cinnamon oil or cinnamaldehyde were added in a range of 0.1 to 0.5%. In these cases, the low content of flavouring did not disturb the network structure.

On the other hand, the effect of relative humidity on flavour transfer rates is probably as important as the effect of temperature. Moisture increases the mass transfer rates of gases and vapours through hydrophilic biopolymer films. Miller, Upadhyaya and Krotchta (1998) observed substantial enhancement of D-limonene permeability through whey protein films when the RH was increased. There was two-fold to 20-fold increase when the RH varied from 40 to 80%. Quezada-Gallo (1999) observed similar behaviour for the permeability of 2-pentanone, 2-heptanone and ethyl esters through both methylcellulose and wheat gluten films. This behaviour was attributed to plasticization of the biopolymer network by water. Although the effect of water on the permeability of volatile compounds is well known, the effect of aroma compounds or essential oils on the WVP is more complex.

Water vapour permeability of soy protein isolate or alginate films (without lipid emulsions) increases when essential oils or aroma compounds were incorporated, whereas films of sodium alginate plus lipid, carrageenan, chitosan or hydroxypropyl methylcellulose decreased. Atarés *et al.* (2010) showed that the incorporation of ginger and cinnamon essential oils into soy protein isolate films resulted in a reduction in the water vapour barrier properties. This fact might be due to possible interactions of oil components with some protein molecules, which could promote a decrease in the hydrophobic character of the protein matrix. The effectiveness of cinnamon oil, as compared to ginger oil, in reducing WVP suggests that the former remains partially integrated in the protein network of the dry films. The difficulties in integrating the essential oils or aroma in a hydrophilic network may be due to matrix disruptions and the creation of void spaces at the protein–essential oil interface. Therefore, it cannot be assumed that the WVP of edible films is reduced simply by adding a hydrophobic component such as an aroma or essential oil to the matrix, but the impact of lipid addition is a determining factor in water barrier efficiency. In the case of hydroxypropyl methylcellulose, chitosan or carrageenan, the WVP increased with the essential oil content. The WVP values showed a significant decrease with the increase in tea tree oil concentration, following a linear trend (Sánchez-González *et al.*, 2010a. 2010b). This behaviour is

expected as an increase in the hydrophobic compound fraction usually leads to an improvement in the water barrier properties of films (Zivanovic, Chiand Draughon, 2005).

12.4.3 Flavour retention and release

The ability of an edible coating to 'capture' or hold on to flavourings during the drying process is critical. If flavours are lost during the drying process, the resultant flavour will lose strength and potentially become unbalanced in character. A secondary concern regarding flavour retention is that a flavour compound not retained in the powder is lost to the processing environment (Reineccius, 2009). Of the common edible coatings used in flavour encapsulation, modified food starches are the best in terms of flavour retention during drying. Modified food starches are excellent emulsifiers, and emulsion quality has a strong influence on flavour retention during spray drying (Baranauskiene *et al.*, 2007; Risch and Reineccius, 1988). Additionally, modified food starches can typically be used at 50–55% solids levels versus 30–35% solids for acacia gum. Thus, both modified food starches and acacia gum will produce good emulsions (i.e. they should yield good flavour retention); modified food starches yield the best flavour retention, because they can be used at a higher solid level, which also improves flavour retention (Reineccius, 1989). Maltodextrins, corn syrup solids or simple sugars (and their alcohols) typically can be used at high solid levels, but their poor emulsification properties result in poor flavour retention during drying. Therefore, a good emulsification capacity of coating material coupled with a high solid content at which they can be used are the major determinant parameters for flavour retention during drying.

Flavour release refers to how an encapsulated flavouring leaves its content. A shortcoming of the major process used for flavour encapsulation (i.e. spray drying) is its inability to use edible coatings that are water-insoluble. The spray drying process requires an aqueous soluble edible coating. It must be used as an encapsulant in order to release flavourings upon contact with water or food product. While, to some extent, all encapsulated flavourings offer controlled release (e.g. release only after contact with water), additional controlled release properties may be desirable in certain product applications (Reineccius, 2009). The main advantages of controlled release are that flavours are released at controlled rates over a prolonged period of time. Hence, the loss of flavourings during processing and even digestive molecular destruction can be strongly reduced (Desobry and Debeaufort, 2012). In the case of an encapsulated flavouring for a dry beverage mix, a rapid release when hydrated is desired. Thus, the desired flavour release will be dependent upon the application. Currently, these controlled release properties can

be attained directly through the use of coacervation and extrusion. Since spray dried particles are water soluble, controlled release properties may be imparted to them through application of secondary coatings, for example fats, oils or gums. Secondary coatings are costly and problematic to apply and therefore it is desirable to accomplish controlled release by choosing an appropriate encapsulation technique.

The release of flavourings can be governed under four different mechanisms individually or in combination: fracturation, diffusion, dissolution or melting, and biodegradation (Barbosa-Cánovas *et al.*, 2005). These are described as follows:

Fracturation. The coating can be fractured or broken open by external forces, such as pressure, shearing or extrasonics. Chewing is the most commonly used mechanical release means.

Diffusion. Given that capsules are very small, they have a large surface area per unit mass. Capsules may function as a semi-permeable membrane, releasing the flavours by a diffusion-controlled process. The slight heat application or increase of solvent concentration (e.g. increase of moisture content) increases permeability by changing the crystalline state of the amorphous matrix into a more mobile rubbery state. Thus, the flow of flavourings through the coating will be facilitated.

Dissolution or *melting*. The integrity of the coating can be destroyed by dissolution into an appropriate solvent or by thermal means. Water-soluble coatings can be easily dissolved from around the core by increasing moisture in the systems. Thermal release is commonly used for fat coatings. In this case, the coating melts away from the core, thus releasing the flavours in an environment such as that occurring during baking.

Biodegradation. Release from coating can be accomplished by biodegradation mechanisms. For example, lipid coatings may be degraded by the action of lipases.

12.4.4 Applications

Flavouring agents can be incorporated directly into edible coatings matrices or can be encapsulated to protect their activity and properties better. Some examples currently used include citric, malic and tartaric acids (Eswaranandam, Hettiarachchy and Johnson, 2006; Eswaranandam, Hettiarachchy and Meullenet, 2006), as well as several essential oils of oregano, thyme, cinnamon, lemongrass and clove that can enhance or mask original flavours of foods. For instance, Rojas-Graü *et al.* (2007a) evaluated the sensory quality of coated fresh-cut 'Fuji' apples containing plant essential oils, such as lemongrass, oregano oil and vanillin incorporated into apple puree–alginate edible coatings. Taste evaluations indicated that coated fresh-cut apples

containing incorporated vanillin (0.3%) were the most promising in terms of sensory quality. Although the use of all these compounds in food has been widely reported, carvacrol (a major component of the essential oils of oregano and thyme) appears to have recently received great attention from researchers. Carvacrol is used as a flavouring agent in baked goods, sweets, ice cream, beverages and chewing gum (Fenaroli, 2009). Moreover, Laohakunjit and Kerdchoechuen (2007) coated milled rice with sorbitol-rice starch coatings containing 25% natural pandan leaf extract (*Pandanus amaryllifolius* Roxb.), allowing the production of jasmine-flavoured rice after cooking.

Several products are already on the market using coating flavouring technology. For instance, there is a roasted peanut with a curry-flavoured coating that is instantaneously dissolved in the mouth and immediately gives the perception of the Indian spice. Another example designed for children is a multi-sugar-coated sweet in which each layer of the coating contains different tastes and flavours, separated by Arabic gum or other hydrocolloid layer(s) to prevent migration of aroma compounds from one layer to another. For this application, diffusivity of volatile compounds should be very low and with a high affinity for the coating, which should be highly soluble in the mouth.

The safety of use of food flavourings, both natural and synthetic, remains however a controversial topic – and will likely elicit debate, motivate scientific studies and entertain legislative actions in the near future.

12.5 Regulatory aspects and future trends

The food industry is perhaps one of the most interesting and difficult industries to regulate. The safety and quality of food products, the way consumers view labelling and intend to use, and processing technologies employed by industries make the decision making difficult.

Initially, and to provide a clear understanding of the food products that are being regulated, it is essential to consider the main specifications of the food product, which requires information about the raw materials and its chemical characterization, and the method of manufacture of the product. Therefore, utilization of flavouring agents and the technique used for its retention and delivery should be clear in order to be regulated.

One of the most evident controversies about flavouring is the flavour definition that depends on the organization that is providing the definition, for example definitions given by the Food and Drug Administration (FDA) and the Council of Europe (CoE). The FDA defines flavours as ‘substances added to impart or help impart a taste or aroma in food’. The CoE defines flavour as: ‘A flavouring substance is a chemically-defined compound which has flavouring properties. It is obtained either by isolation from a natural source

or by synthesis. Flavouring properties are those which are predominantly odour-producing and which may also affect the taste'. On the other hand, the Food and Agriculture Organization of the United Nations and the World Health Organization (WHO/FAO) Expert Committee on Food Additives and Contaminants (JECFA) do not have a definition of what constitutes an artificial or natural flavour (Fenaroli, 2009; FAO, 2005a).

Worldwide, in European countries and in countries like the United States of America (USA), Canada and Australia, food regulation has already existed for decades (Salzer, 2007). Aiming at a global regulation, FAO have their own guidelines trying to make the food regulation in all countries uniform. Created in 1957 by the FAO and the WHO, the JECFA have a mission on the evaluation of food additives where flavours are considered. Flavours are included in Addendum 13 of the Combined Compendium of Food Additive Specifications document (FAO, 2005b) and in a new document published in 2012 by the FAO (2012), where new specifications for certain flavourings were revised and new specifications were added. In 2008, JECFA has evaluated more than 1750 substances and this list is updated annually. Based on these conclusions, JECFA in collaboration with the Flavour and Extract Manufacturers Association (FEMA) have created an open positive list of flavouring substances for use in the USA and in WHO member countries that wish to adopt the FEMA and/or JECFA lists. Also, in the European Union (EU), a positive list is being compiled based on the safety evaluations performed by JECFA prior to 2000 and thereafter by the European Food Safety Authority (EFSA) (Smith *et al.*, 2009).

Besides FAO documents, guidelines provided by the CODEX Alimentarius are also used by some countries. In 2005, the CODEX Alimentarius Commission presented a discussion paper concerning the development of guidelines for flavouring agents. In this document, flavour definition is given as: 'Flavour is the sum of those characteristics of any material taken in the mouth, perceived principally by the senses of taste and smell, and also the general pain and tactile receptors in the mouth, as received and interpreted by the brain. The perception of flavour is a property of flavourings.' They presented the guidelines providing the principles for the safe use of flavourings (CODEX-Alimentarius, 2005).

One the most amazing differences in regulations is the fact that in the USA flavours, regulated by the FDA, are food additives, while in Europe, where they are regulated by the European Community and the EFSA, they are treated differently with one separated directive. Food additives are regulated by a list where all the substances that can be used are presented. In some countries there is a list of substances that cannot be used (Salzer, 2007). In the EU, a food additive can be used in food if the EFSA has found that it is harmless to the health of consumers and that it has an established ADI (acceptable daily intake). Besides this, there must be a technological need for the utilization of the substance (Salzer, 2007). Recently, and following a

request from the Commission, the Scientific Panel on Food Contact Materials, Enzymes, Flavourings and Processing Aids, from EFSA, provided scientific advice regarding the data required for the evaluation of flavourings. In Part A of these guidelines a proposal was presented concerning the data required for risk assessment of flavouring substances, that is chemically defined substances with flavouring properties. Part B of these guidelines provides a proposal concerning the data required for risk assessment. It categories flavourings for which an evaluation and an approval is required. These are Regulation (EC) No. 1334/2008 of the European Parliament and of the Council on flavourings (EFSA, 2010; Regulation (EC) No. 1334/2008, 2008).

The FDA presents, together with the International Food Information Council (IFIC), the regulation aspects of food ingredients and colours (including flavours) in additives. They present an explanatory document in order to determine the regulatory status of a food ingredient and the way they should be evaluated. They say that any substance that is reasonably expected to become a component of food is a food additive that is subject to pre-market approval by the FDA, unless the substance is generally recognized as safe (GRAS) among experts qualified by scientific training and experience to evaluate its safety under the conditions of its intended use or meets one of the other exclusions from the food additive definition of the Federal Food, Drug, and Cosmetic Act (FFDCA) (FDA, 2012).

Besides all these regulatory aspects, there are two nongovernmental associations that present very clear documents about flavouring and flavour agents. Since 2009, the European Flavour Association (EFFA) changes its scope to focus only on flavourings, as their members are essentially flavouring companies that manufacture or blend flavours within the countries of the European Economic Area, but also including other national associations from across the European Union (EFFA, 2012). The International Organization of the Flavour Industry (IOFI) also tries to provide updated information about flavouring to their members. The IOFI Code of Practice is a document that guides all its members, being a global platform for the production and sale of safe materials to be used as flavour ingredients (IOFI, 2012).

The process for a new flavour admission is very expensive and long, which normally impedes development and may result in the abandonment of flavours registration, mainly due to current safety regulations and toxicity studies that in these cases are mandatory (Regnier, Combrinck and Du Plooy, 2012). EFSA in 2012 presented guidance for the submission of a food additive evaluation request, where flavours are included, and the applications for authorization of a new food additive or to the modification of an already authorized food additive. Data requirements and their context and the risk assessment paradigm applied are described (EFSA, 2012). Also the Regulation (EC) No. 1331/2008 established the procedures for a common authorization for food additives, food enzymes and food flavourings, and should be used as a guide for new products (Regulation (EC) No. 1331/2008).

The utilization of flavouring agents is reported in several studies and in different food products; they can be used with the intention of flavouring but they also can be used as natural food preservatives, due to their antibacterial and antioxidant activities, protecting food against well-known causal agents of food-borne diseases and food spoilage. However, to further confirm the safety of food flavours, studies on their safety issues must be conducted. Also the processing of food using coating technology for preservation and flavouring should be explored, as these coatings were shown to have a minimal effect on the nutritional quality of foods, being an interesting way to add functionality to food products without changing their properties.

More studies have to be performed in order to understand how the retention and release of flavours behave in edible-coating matrices and if nanotechnology and the utilization of nanostructured materials could lead to improved properties when compared with macro- and microstructures.

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References

Abbas, S., Da Wei, C., Hayat, K. and Xiaoming, Z. (2012) Ascorbic acid: microencapsulation techniques and trends: a review. *Food Reviews International*, **28**, 343–374.

Abdollahi, A., Hassani, A., Ghosta, Y., Bernousi, I., Meshkatalsadat, M.H., Shabani, R. and Ziaee, S.M. (2012) Evaluation of essential oils for maintaining post harvest quality of Thompson seedless table grape. *Natural Product Research*, **26**, 77–83.

Abegaz, E.G., Tandon, K.S., Scott, J.W., Baldwin, E.A. and Shewfelt, R.L. (2004) Partitioning taste from aromatic flavour notes of fresh tomato (*Lycopersicon esculentum*, Mill) to develop predictive models as a function of volatile and monovolatile components. *Postharvest Biology and Technology*, **34**, 227–235.

Albarracín, W., Alfonso, C. and Sanchez, I.C. (2012) Application of essential oils as a preservative to improve the shelf life of Nile tilapia (*Oreochoromisniloticus*). *Vitae, Revista da Faculdade de Química Farmacêutica*, **19**, 34–40.

Al-Reza, S.M., Rahman, A., Lee, J. and Kang, S.C, (2010) Potential roles of essential oil and organic extracts of *Zizyphus jujuba* in inhibiting food-borne pathogens. *Food Chemistry*, **119**, 981–986.

Amefia, A.E., Abu-Ali, J.M. and Barringer, S.A. (2006) Improved functionality of food additives with electrostatic coating. *Innovative Food Science and Emerging Technologies*, **7**, 176–181.

Anker, M., Berntsen J., Hermansson, A.M. and Stading, M. (2002) Improved water vapor barrier of whey protein films by addition of an acetylated monoglyceride. *Innovative Food Science and Emerging Technologies*, **3**, 81–92.

Antoun, N. and Tsimidou, M. (1997) Gourmet olive oils: stability and consumer acceptability studies. *Food Research International*, **30**, 131–136.

ASTM (1995) *Standard test methods for water vapor transmission of material.* In: *Annual Book of ASTM*. American Society for Testing and Materials, Philadelphia, Pennsylvania, USA, pp. E96–95.

Atarés, L., De Jesús, C., Talens, P. and Chiralt, A. (2010) Characterization of SPI-based edible films incorporated with cinnamon or ginger essential oils. *Journal of Food Engineering*, **99**, 384–391.

Attokaran, M. (2011) Flavors. In: *Natural Food Flavors and Colorants*. Wiley-Blackwell, pp. 9–10.

Bai, J., Alleyne, V., Hagenmaier, R.D., Mattheis, J.P. and Baldwin, E.A. (2003) Formulation of zein coatings for apples (*Malus domestica*). *Postharvest Biology and Technology*, **28**, 259–268.

Bakkali, F., Averbeck, S., Averbeck, D. and Idaomar, M. (2008) Biological effects of essential oils – a review. *Food and Chemical Toxicology*, **46**, 446–475.

Baldwin, E.A., Nispero-Carriedo, M.O., Hagenmaier, R.D. and Baker, R.A. (1997) Use of lipids in coatings for food products. *Food Technology*, **51**, 56–64.

Baranauskiene, R., Bylaite, E., Zukauskaite, J. and Venskutonis, R.P. (2007) Flavor retention of peppermint (*Mentha piperita* L.) essential oil spray-dried in modified starches during encapsulation and storage. *Journal of Agricultural and Food Chemistry*, **55**, 3027–3036.

Barbosa-Cánovas, G.V., Ortega-Rivas, E., Yan, H. and Juliano P. (2005) Encapsulation processes. In: *Food Powders: Physical Properties, Processing, and Functionality*. Food Engineering Series, Kluwer Academic/Plenum Publishers, New York, USA, pp. 199–219.

Beirão da Costa, S., Duarte, C., Bourbon, A.I., Pinheiro, A.C., Serra, A.T., Moldão Martins, M., Nunes Januário, M.I., Vicente, A.A., Delgadillo, I., Duarte, C. and Beirão da Costa, M.L. (2012) Effect of the matrix system in the delivery and *in vitro* bioactivity of microencapsulated oregano essential oil. *Journal of Food Engineering*, **110**, 190–199.

Belletti, N., Kamdem, S.S., Tabanelli, G., Lanciotti, R. and Gardini, F. (2010) Modeling of combined effects of citral, linalool and β-pinene used against *Saccharomyces cerevisiae* in citrus-based beverages subjected to a mild heat treatment. *International Journal of Food Microbiology*, **136**, 283–289.

Belščak-Cvitanović, A., Stojanović, R., Manojlović, V., Komes, D., Juranović Cindrić, I., Nedović, V. and Bugarski, B. (2011) Encapsulation of polyphenolic antioxidants from medicinal plant extracts in alginate–chitosan system enhanced with ascorbic acid by electrostatic extrusion. *Food Research International*, **44**(4), 1094–1101.

Beristain, C.I., García, H.S. and Vernon-Carter, E.J. (1999) Note. Mesquite gum (*Prosopis juliflora*) and maltodextrin blends as wall materials for spray-dried encapsulated orange peel oil. *Food Science and Technology International*, **5**, 353–356.

Beristain, C.I., García, H.S. and Vernon-Carter, E.J. (2001) Spray-dried encapsulation of cardamom (*Elettaria cardamomum*) essential oil with mesquite (*Prosopis juliflora*) gum. *LWT-Food Science and Technology*, **34**(6), 398–401.

Bierhalz, A., Altenhofen da Silva, M. and Kieckbusch, T. (2012) Natamycin release from alginate/pectin films for food packaging applications. *Journal of Food Engineering*, **110**, 18–25.

Bifani, V., Ramírez, C., Ihla, M., Rubilara, M., García, M.A. and Zaritzky, N. (2007) Effects of murta (*Ugni molinae* Turcz) extract on gas and water vapor permeability of carboxymethylcellulose based edible films. *LWT-Food Science and Technology*, **40**, 1473–1481.

Bonvehí, J.S. (2005) Investigation of aromatic compounds in roasted cocoa powder. *European Food Research and Technology*, **221**, 19–29.

Bourbon, A.I., Pinheiro, A.C., Cerqueira, M.A., Rocha, C.M., Avides, M.C., Quintas, M.A. and Vicente A.A. (2011) Physico-chemical characterization of chitosan-based edible films incorporating bioactive compounds of different molecular weight. *Journal of Food Engineering*, **106**, 111–118.

Bravin, B., Peressini, D. and Sensidoni, A. (2006) Development and application of polysaccharide–lipid edible coating to extend shelf-life of dry bakery products. *Journal of Food Engineering*, **76**, 280–290.

Buonocore, G.G., Del Nobile, M.A., Di Martino, C., Gambacorta, G., La Notte, E. and Nicolais, L. (2003) Modeling the water transport properties of casein-based edible coating. *Journal of Food Engineering*, **60**, 99–106.

Burt, S. (2004) Essential oils: their antibacterial properties and potential applications in foods: a review. *International Journal of Food Microbiology*, **94**, 223–253.

Byun, Y., Kim, T., Desai, K.G.H. and Park, H.J. (2010) Microencapsulation techniques for food flavour. In: *Chemistry and Biology of Volatiles* (Herrmann, A., ed.). John Wiley & Sons, Ltd, pp. 307–332.

Cabral, L.M.C., Bravo, A., Freire, M., Bizzo, H.R. and Matta, V.M. (2010) Citrus fruits and oranges. In: *Handbook of Fruit and Vegetable Flavors* (Hui, Y.H., ed.). John Wiley & Sons, Inc., pp. 265–278.

Cerqueira, M.A. (2010) Development and characterization of edible coatings to the preservation of cheese quality. PhD Thesis, University of Minho, Portugal.

Cerqueira, M.A., Lima, A.M., Souza, B.W.S., Teixeira, J.A., Moreira, R.A. and Vicente, A.A. (2009) Functional polysaccharides as edible coatings for cheese. *Journal of Agricultural and Food Chemistry*, **57**, 1456–1462.

Cerqueira, M.A, Sousa-Gallagher, M.J., Macedo, I., Rodriguez-Aguilera, R., Souza, B.W.S., Teixeira, J.A. and Vicente, A.A. (2010a) Use of galactomannan edible coating application and storage temperature for prolonging shelf-life of 'Regional' cheese. *Journal of Food Engineering*, **97**, 87–94.

Cerqueira, M.A., Souza, B.W.S., Martins, J.T., Teixeira, J.A. and Vicente, A.A. (2010b) Seed extracts of *Gleditsia triacanthos*: Functional properties evaluation and incorporation into galactomannan films. *Food Research International*, **43**, 2031–2038.

Cerqueira, M.A., Bourbon, A.I., Pinheiro, A.C., Martins, J.T., Souza, B.W.S., Teixeira, J.A. and Vicente, A.A. (2011a) Galactomannans use in the development of edible

films/coatings for food applications. *Trends in Food Science and Technology*, **22**, 662–671.

Cerqueira, M.A., Souza, B.W.S., Simões, J., Teixeira, J.A., Domingues, M.R., Coimbra, M.A. and Vicente, A.A. (2011b) Structural and thermal characterization of galactomannans from non-conventional sources. *Carbohydrate Polymers*, **83**, 179–185.

Cerqueira, M.A., Souza, B.W.S., Teixeira, J.A. and Vicente, A.A. (2012) Effect of glycerol and corn oil on physicochemical properties of polysaccharide films: a comparative study. *Food Hydrocolloids*, **27**, 175–184.

Cervera, M.F., Karjalainen, M., Airaksinen, S., Rantanen, J., Krogars, K., Heinämäki, J., Colarte, A.I. and Yliruusi, J. (2004) Physical stability and moisture sorption of aqueous chitosan–amylose starch films plasticized with polyols. *European Journal of Pharmaceutics and Biopharmaceutics*, **58**, 69–76.

Che Man, Y.B., Irwandi, J. and Abdullah, W.J. (1999) Effect of different types of maltodextrin and drying methods on physico-chemical and sensory properties of encapsulated durian flavor. *Journal of the Science of Food and Agriculture*, **79**, 1075–1080.

Chiumarelli, M. and Hubinger, M.D. (2012) Stability, solubility, mechanical and barrier properties of cassava starch and carnauba wax edible coatings to preserve fresh-cut apples. *Food Hydrocolloids*, **28**, 59–67.

Clark, J.E. (1998) Taste and flavour: their importance in food choice and acceptance. *Proceedings of the Nutrition Society*, **57**, 639–643.

Cleveland, J., Montville, T.J., Nes, I.F. and Chikindas, M.L. (2001) Bacteriocins: safe, natural antimicrobials for food preservation. *International Journal of Food Microbiology*, **71**, 1–20.

CODEX Alimentarius (2005) CX/FAC 06/38/12 December 2005, Joint FAO/WHO Food Standards Programme.

CODEX Alimentarius (2012) List of Codex Specifications for Food Additives. CAC/MISC 6-2012. http://www.codexalimentarius.org/download/standards/9/CXA_006e.pdf.

Cuq, B., Gontard, N. and Guilbert, S. (1995) Edible films and coatings as active layers. In: *Active Food Packaging* (Rooney, M., ed.). Blackie Academic and Professional, Glasgow, Scotland, pp. 111–142.

Cuq, B., Gontard, N., Cuq, J.L., and Guilber, S. (1996) Functional properties of myofibrillar protein-based biopackaging as affected by film thickness. *Journal of Food Science*, **61**, 580–584.

Cutter, C.N. (2000) Antimicrobial effect of herb extracts against *Escherichia coli* O157:H7, *Listeria monocytogenes*, and *Salmonella typhimurium* associated with beef. *Journal of Food Protection*, **63**, 601–607.

Cutter, C.N. and Sumner, S.S. (2002) Application of edible coatings on muscle foods. In: *Protein-Based Films and Coatings* (Gennadios, A., ed.). CRC Press, Boca Raton, Florida, USA, pp. 467–484.

Debeaufort, F. and Voilley, A. (2009) Lipid-based edible films and coatings. In: *Edible Films and Coatings for Food Applications* (Embuscado, M. and Huber, K.C., eds). Springer, New York, USA, pp. 135–168.

de Roos, K.B. (2003) Effect of texture and microstructure on flavour retention and release. *International Dairy Journal*, **13**, 593–605.

Desai, K.G.H. and Park, J.H. (2005a) Recent developments in microencapsulation of food ingredients. *Drying Technology*, **23**, 1361–1394.

Desai, K.G.H. and Park, H.J. (2005b) Encapsulation of vitamin C in tripolyphosphate cross-linked chitosan microspheres by spray drying. *Journal of Microencapsulation*, **22**(2), 179–192.

Desobry, S. and Debeaufort, F. (2012) Encapsulation of flavors, nutraceuticals, and antibacterials. In: *Edible Coatings and Films to Improve Food Quality* (Baldwin, E.A., ed.). CRC Press, Boca Raton, Florida, USA, pp. 333–372.

de Vos, P., Faas, M.M., Spasojevic, M. and Sikkema, J. (2010) Encapsulation for preservation of functionality and targeted delivery of bioactive food components. *International Dairy Journal*, **20**, 292–302.

Dorman, H.J.D. and Deans, S.G. (2000) Antimicrobial agents from plants: antibacterial activity of plant volatile oils. *Journal of Applied Microbiology*, **88**, 308–316.

Druaux, C. and Voilley, A. (1997) Effect of food composition and microstructure on volatile flavour release. *Trends in Food Science and Technology*, **8**, 364–368.

Drusch, S., Serfert, Y., Scampicchio, M., Schmidt-Hansberg, B. and Schwarz, K. (2007) Impact of physicochemical characteristics on the oxidative stability of fish oil microencapsulated by spray-drying. *Journal of Agricultural and Food Chemistry*, **55**, 11044–11051.

EFFA (2012) European Flavour Association. http://www.effa.eu/en/about/welcome-to-effa. Accessed 11.11.2012.

EFSA (2010) Panel on Food Contact Materials, Enzymes, Flavourings and Processing Aids; Draft Guidance on the data required for the risk assessment of flavourings. *EFSA Journal*, **8**(6), 1623 [38 pp.]. DOI:10.2093/j.efsa.2010.1623. Available online: www.efsa.europa.eu.

EFSA (2012) Panel on Food Additives and Nutrient Sources Added to Food (ANS); Guidance for submission for food additive evaluations. *EFSA Journal*, **10**(7), 2760 [60 pp.]. DOI: 10.2903/j.efsa.2012.2760. Available online at: www.efsa.europa.eu/efsajournal.

Elayedath, S. and Barringer, S.A. (2002) Electrostatic powder coating of shredded cheese with antimycotic and anticaking agents. *Innovative Food Science and Emerging Technologies*, **3**, 385–390.

El-Ghorab, A., El-Massry, K.F. and Shibamoto, T. (2007) Chemical composition volatile extract and antioxidant activities of the volatile and nonvolatile extracts of Egyptian corn silk (*Zea mays* L.). *Journal of Agricultural and Food Chemistry*, **55**, 9124–9127.

Erkan, N., Ayranci, G. and Ayranci, E. (2008) Antioxidant activities of rosemary (*Rosmarinus officinalis* L.) extract, blackseed (*Nigella sativa* L.) essential oil, carnosic acid, rosmarinic acid and sesamol. *Food Chemistry*, **110**, 76–82.

Eswaranandam, S., Hettiarachchy, N.S. and Johnson, M.G. (2006a) Antimicrobial activity of citric, lactic, malic, or tartaric acids and nisin-incorporated soy protein film against *Listeria monocytogenes, Escherichia coli* O157:H7, and *Salmonella gaminara*. *Journal of Food Science*, **69**, 79–84.

Eswaranandam, S., Hettiarachchy, N. S. and Meullenet, J.F. (2006b) Effect of malic and lactic acid incorporated soy protein coatings on the sensory attributes of whole apple and fresh-cut cantaloupe. *Journal of Food Science*, **71**, 307–313.

Evrendilek, G.A. and Balasubramaniam, V.M. (2011) Inactivation of *Listeria monocytogenes* and *Listeria innocua* in yogurt drink applying combination of high pressure processing and mint essential oils. *Food Control*, **22**, 1435–1441.

Fadini, A.L., Rocha, F.S., Alvima, I.D. Sadahira, M.S. Queiroz, M.B., Alves, R.M.V. and Silva, L.B. (2013) Mechanical properties and water vapor permeability of hydrolysed

collagene–cocoa butter edible films plasticized with sucrose. *Food Hydrocolloids*, **30**, 625–631.

Fajardo, P., Martins, J.T., Fuciños, C., Pastrana, L., Teixeira, J.A. and Vicente A.A (2010) Evaluation of a chitosan-based edible film as carrier of natamycin to improve the storability of Saloio cheese. *Journal of Food Engineering*, **101**, 349–356.

Fang, Z., and Bhandari, B. (2012) Encapsulation techniques for food ingredient systems. In: *Food Materials Science and Engineering* (Bhandari, B. and Roos, Y.H., eds). Wiley-Blackwell, pp. 320–348.

FAO (2005a) Combined Compendium of Food Additive Specifications: Joint FAO/WHO Expert Committee on Food Additives: All Specifications Monographs from the 1st to the 65th Meeting (1956–2005). http://www.fao.org/docrep/009/a0691e/a0691e00.htm.

FAO (2005b) Combined Compendium of Food Additive Specifications: Joint FAO/WHO Expert Committee on Food Additives: All Specifications Monographs from the 1st to the 65th Meeting (1956-2005), Addendum **13**.

FAO (2012) Combined Compendium of Food Additive Specifications: Joint FAO/WHO Expert Committee on Food Additives. 76th Meeting 2010, Addendum 13.

FDA (2012) http://www.fda.gov/Food/FoodIngredientsPackaging/FoodAdditives/ucm 228269.htm. Accessed 11.11.2012.

Fellows, P. (2000) Coating or enrobing. In: *Food Processing Technology: Principles and Practice* (Fellows, P., ed.), 3rd edn. Woodhead, Cambridge, England, pp. 455–461.

Fenaroli, G. (ed.) (2009) *Fenaroli's Handbook of Flavor Ingredients*, 6th edition. CRC Press, Boca Raton, Florida, USA.

Flores, M., Aristoy, M.C., Spanier, A.M. and Toldrá, F. (1997) Non-volatile components effects on quality of 'Serrano' dry-cured ham as related to processing time. *Journal of Food Science*, **62**, 1235–1239.

Frascareli, E.C., Silva, V.M., Tonon, R.V. and Hubinger, M.D. (2012) Effect of process conditions on the microencapsulation of coffee oil by spray drying. *Food and Bioproducts Processing*, **90**(3), 413–424.

García, M.A., Martino, M.N. and Zaritzky, N.E. (1999) Edible starch films and coatings characterization: scanning electron microscopy, water vapor, and gas permeabilities. *Scanning*, **21**, 348–353.

García, M.A., Martino, M.N. and Zaritzky, N.E. (2000) Microstructural characterization of plasticized starch-based films. *Starch/Staerke*, **52**, 118–124.

García, M.A., Pinotti, A. and Zaritzky, N.E. (2006) Physicochemical, water vapor barrier and mechanical properties of corn starch and chitosan composite films. *Starch/Staerke*, **58**, 453–463.

García, M.A., Pinotti, A., Martino, M.N. and Zaritzky, N.E. (2004) Characterization of composite hydrocolloid films. *Carbohydrate Polymers*, **56**, 339–345.

García, L.C., Pereira, L.M., Sarantópoulos, C.I.G.L. and Hubinger, M.D. (2012) Effect of antimicrobial starch edible coating on shelf-life of fresh strawberries. *Packaging Technology and Science*, **25**, 413–425.

Gennadios, A., McHugh, T.H., Weller, C.L. and Krochta J.M. (1994) Edible coatings and film based proteins. In: *Edible Coatings and Films to Improve Food Quality* (Krochta, J.M., Baldwin, E.A., Nisperos-Carriedo, M., eds). Technomic Publishing, Lancaster, Pennsylvania, pp. 201–277.

Gharsallaoui, A., Roudaut, G., Chambin, O., Voilley, A. and Saurel, R. (2007) Applications of spray-drying in microencapsulation of food ingredients: an overview. *Food Research International*, **40**, 1107–1121.

Gilleo, K.B. (2007) Rheology and surface chemistry. In: *Coatings Technology: Fundamentals, Testing and Processing Techniques* (Tracton, A.A., ed.). Taylor & Francis Group, pp. 1–12.

Gniewosz, M. and Synowiec, A. (2011) Antibacterial activity of pullulan films containing thymol. *Flavour and Fragrance Journal*, **26**, 389–395.

González, L., Vargas, M., González-Martínez, C., Chiralt, A. and Cháfer, M. (2011) Use of essential oils in bioactive edible coatings: a review. *Food Engineering Reviews*, **3**, 1–16.

Gouin, S. (2004) Microencapsulation: industrial appraisal of existing technologies and trends. *Trends in Food Science and Technology*, **15**, 330–347.

Gould, G.W. (2000) Preservation: past, present and future. *British Medical Bulletin*, **56**, 84–96.

Grandison, A.S. (2006) Postharvest handling and preparation of foods for processing. In: *Food Processing Handbook* (Brennan, J.G., e.d). Wiley-VCH Verlag GmbH & Co., KGaA, Weinheim, Germany, pp. 1–30.

Grant, L. and Burns, J.K. (1994) Application of coatings. In: *Edible Coatings and Films to Improve Food Quality* (Krochta, J.M., Baldwin, E.A. and Nisperos-Carriedo, M., eds). Technomic, Lancaster, Pennsylvania, USA, pp. 189–200.

Guangrong, H., Jiaxin, J. and Dehui, D. (2008) Antioxidative and antibacterial activity of the methanol extract of *Artemisia anomala* S. Moore. *African Journal of Biotechnology*, **7**, 1335–1338.

Guilbert, S. (1986) Technology and application of edible protective films. In: *Food Packaging and Preservation. Theory and Practice* (Mathlouthi, M., ed.). Elsevier Applied Science Publishing Co., London, England, pp. 371–394.

Guilbert, S. and Gontard, N. (1995) Edible and biodegradable food packaging. In: *Foods and Packaging Materials – Chemical Interactions* (Ackermann, P., Jagerstad, M. and Ohlsson, T., eds). The Royal Society of Chemistry, Cambridge, England, pp. 159–168.

Guilbert, S. and Gontard, N. (2005) Agro-polymers for edible and biodegradable films: review of agricultural polymeric materials, physical and mechanical characteristics. In: *Innovations in Food Packaging* (Han, J.H., ed.). Elsevier Academic, Oxford, England, pp. 263–276.

Guilbert, S., Gontard, N. and Gorris, L.G.M. (1996) Prolongation of the shelf-life of perishable food products using biodegradable films and coatings. *LWT-Food Science and Technology*, **29**, 10–17.

Gupta, V. and Nair, J. (2012) Application of botanicals as natural preservatives in food. In: *Progress in Food Preservation* (Bhat, R., Alias, A.K. and Paliyath, G., eds). Wiley-Blackwell, pp. 513–530.

Hamad, S.H. (2012) Factors affecting the growth of microorganisms in food. In: *Progress in Food Preservation* (Bhat, R., Alias, A.K. and Paliyath, G., eds). Wiley-Blackwell, pp. 405–427.

Hambleton, A., Debeaufort, F., Beney, L., Karbowiak, T. and Voilley, A. (2008) Protection of active aroma compound against moisture and oxygen by encapsulation in biopolymeric emulsion-based edible films. *Biomacromolecules*, **9**(3), 1058–1063.

Hambleton, A., Fabra, M. J., Debeaufort, F., Brun-Dury, C. and Voilley, A. (2009) Interface and aroma barrier properties of iota-carrageenan emulsion-based films used for encapsulation of active food compounds. *Journal of Food Engineering*, **93**, 80–88.

Hambleton, A., Perpiñan-Saiz, N., Fabra, M.J., Voilley, A. and Debeaufort, F. (2012) The Schroeder paradox or how the state of water affects the moisture transfers through edible films. *Food Chemistry*, **132**(4), 1671–1678.

Han, J. (2002) Protein-based edible films and coatings carrying antimicrobial agents. In: *Protein-Based Films and Coatings* (Gennadios, A., ed.). CRC, Boca Raton, Florida, USA, pp. 485–498.

Han, J.H. and Gennadios, A. (2005) Edible films and coatings: a review. In: *Innovations in Food Packaging* (Han, J.H., ed.). Elsevier Academic, Oxford, England, pp. 239–259.

Han, C., Zhao, Y., Leonard, S.W. and Traber, M.G. (2004) Edible coatings to improve storability and enhance nutritional value of fresh and frozen strawberries (*Fragaria ananassa*) and raspberries (*Rubus ideaus*). *Postharvest Biology and Technology*, **33**, 67–78.

Hyldgaard, M., Mygind, T. and Meyer, R.L. (2012) Essential oils in food preservation: mode of action, synergies, and interactions with food matrix components. *Frontiers in Microbiology*, **3**, 1–24.

Inouye, S., Takizawa, T. and Yamaguchi, H. (2001) Antibacterial activity of essential oils and their major constituents against respiratory tract pathogens by gaseous contact. *Journal of Antimicrobial Chemotherapy*, **47**, 565–573.

IOFI (2012) International Organization of the Flavor Industry. http://www.iofi.org/. Accessed 13.11.2012.

Jafari, M.S., Assadpoor, E., He, Y. and Bhandari, B. (2008) Encapsulation efficiency of food flavours and oils during spray drying. *Drying Technology: An International Journal*, **26**(7), 816–835.

Jiang, J., Xiong, Y.L. Newman, M.C. and Rentfrow, G.K. (2012) Structure-modifying alkaline and acidic pH-shifting processes promote film formation of soy proteins. *Food Chemistry*, **132**, 1944–1950.

Juneja, V.K., Dwivedi, H.P. and Yan, X. (2012) Novel natural food antimicrobials. *Annual Review of Food Science and Technology*, **3**, 381–403.

Kadam, M.L., Hashmi, S.I. and Kale, R.V. (2011) Studies on extraction of ginger oil and its microencapsulation. *Electronic Journal of Environmental, Agricultural and Food Chemistry*, **10**(6), 2382–2390.

Karbowiak, T., Debeaufort, F. and Voilley, A. (2006) Importance of surface tension characterization for food, pharmaceutical and packaging products: a review. *Critical Reviews in Food Science and Nutrition*, **46**, 391–407.

Kester, J.J. and Fennema, O. (1986) Edible films and coatings: a review. *Food Technology*, **40**, 47–59.

Khan, M.K.I., Schutyser, M.A.I., Schroën, K. and Boom, R.M. (2012) Electrostatic powder coating of foods – State of the art and opportunities. *Journal of Food Engineering*, **111**, 1–5.

Knights, J. (2010) Flavour legislation. In: *Food Flavour Technology* (Taylor, A.J. and Linforth, R., eds). Wiley-Blackwell, pp. 24–50.

Koehler, P., Kieffer, R. and Wieser, H. (2010) Effect of hydrostatic pressure and temperature on the chemical and functional properties of wheat gluten. III. Studies on gluten films. *Journal of Cereal Science*, **51**,140–145.

Kralovec, J.A., Zhang, S., Zhang, W. and Barrow, C.J. (2012) A review of the progress in enzymatic concentration and microencapsulation of omega-3 rich oil from fish and microbial sources. *Food Chemistry*, **131**, 639–644.

Krishnan, S., Kshirsagar, A.C. and Singhal, R.S. (2005) The use of gum Arabic and modified starch in the microencapsulation of a food flavoring agent. *Carbohydrate Polymers*, **62**, 309–315.

Krochta, J.M. (1997) Edible protein films and coatings. In: *Food Proteins and Their Applications* (Damodaran, S. and Paraf, A., eds). Marcel Dekker, New York, USA, pp. 529–549.

Krochta, J.M. (2002) Proteins as raw materials for films and coatings: definitions, current status, and opportunities. In: *Protein-Based Films and Coatings* (Gennadios, A., ed.). CRC, Boca Raton, Florida, USA, pp. 1–41.

Lakkis, J.M. (2007) Microencapsulation of flavors by complex coacervation, In: *Encapsulation and Controlled Release Technologies in Food Systems* (Thies, C., ed.), Chapter 7. Blackwell Publishing, pp. 149–170.

Laohakunjit, N. and Kerdchoecuen, O. (2007) Aroma enrichment and the change during storage of non-aromatic milled rice coated with extracted natural flavor. *Food Chemistry*, **101**, 339–344.

Lee, Y.-W. and Lee, Y.-Y. (2003) Flavor and aroma substances. In: *Extraction Optimization in Food Engineering* (Liadakis, G. and Tzia, C., eds), Chapter 9. CRC Press, Boca Raton, Florida, USA.

Lima, A.M., Cerqueira, M.A., Souza, B.W.S., Santos, E.C.M., Teixeira, J.A., Moreira, R.A. and Vicente, A.A. (2010) New edible coatings composed of galactomannans and collagen blends to improve the postharvest quality of fruits – influence on fruits gas transfer rate. *Journal of Food Engineering*, **97**, 101–109.

Longo, M.A. and Sanromán, M.A. (2010) Vegetable flavors from cell culture. In: *Handbook of Fruit and Vegetable Flavors* (Hui, Y.H., ed.). John Wiley & Sons, Inc., pp. 663–680.

Lopez, O.V., García, M.A. and Zaritzky, N. (2008) Film forming capacity of chemically modified corn starches. *Carbohydrate Polymers*, **73**, 573–581.

Madene, A., Jacquot, M., Scher, J. and Desobry, S. (2006) Flavour encapsulation and controlled release – a review. *International Journal of Food Science and Technology*, **41**, 1–21.

Mali, S., Grossmann, M.V.E., García, M.A., Martino, M.N. and Zaritzky, N.E. (2005) Mechanical and thermal properties of yam starch films. *Food Hydrocolloids*, **19**, 157–164.

Mali, S., Grossmann, M.V.E., García, M.A., Martino, M.N. and Zaritzky, N.E. (2006) Effects of controlled storage on thermal, mechanical and barrier properties of plasticized films from different starch sources. *Journal of Food Engineering*, **75**, 453–460.

Manojlovic, V., Rajic, N., Djonlagic, J., Obradovic, B., Nedovic, V. and Bugarski, B. (2008) Application of electrostatic extrusion – flavour encapsulation and controlled release. *Sensors*, **8**, 1488–1496.

Martín-Belloso, O., Rojas-Graü, M.A. and Soliva-Fortuny, R. (2009) Delivery of flavour and active ingredients using edible films and coatings. In: *Edible Films and Coatings for Food Applications* (Embuscado, M.E. and Huber K.C., eds), Chapter 10. Springer, New York, USA.

Martins, J.T, Cerqueira, M.A. and Vicente, A.A. (2012) Influence of α-tocopherol on physicochemical properties of chitosan-based films. *Food Hydrocolloids*, **27**, 220–227.

Martins, J.T., Cerqueira, M.A., Souza, B.W.S., Avides, M.C..and Vicente, A.A. (2010) Shelf life extension of ricotta cheese using coatings of galactomannans from nonconventional sources incorporating nisin against *Listeria monocytogenes*. *Journal of Agricultural and Food Chemistry*, **58**, 1884–1891.

Martins, J.T., Cerqueira, M.A., Bourbon, A.I., Pinheiro, A.C., Souza, B.W.S. and Vicente, A.A. (2012) Synergistic effects between κ-carrageenan and locust bean gum on physicochemical properties of edible films made thereof. *Food Hydrocolloids*, **29**, 280–289.

Martin-Sánchez, A.M., Chaves-López, C., Sendra, E., Sayas, E., Fernández-López, J. and Pérez-Álvarez, J.Á. (2011) Lipolysis, proteolysis and sensory characteristics of a Spanish fermented dry-cured meat product (salchichón) with oregano essential oil used as surface mold inhibitor. *Meat Science*, **89**, 35–44.

Mastromatteo M., Conte, A. and Del Nobile, M.A. (2012) Packaging strategies to prolong the shelf life of fresh carrots (*Daucus carota* L.). *Innovative Food Science and Emerging Technologies*, **13**, 215–220.

McHugh, T.H. and Krochta, J.M. (1994) Permeability properties of edible films. In: *Edible Coatings and Films to Improve Food Quality* (Krochta, J.M. Baldwin, E.A. and Nisperos-Carriedo, M., eds). Technomic, Lancaster, Pennsylvalia, USA, pp. 139–187.

Mejlholm, O. and Dalgaard, P. (2002) Antimicrobial effect of essential oils on the seafood spoilage micro-organism *Photobacterium phosphoreum* in liquid media and fish products. *Letters in Applied Microbiology*, **34**, 27–31.

Miller, K.S. and Krochta, J.M. (1997) Oxygen and aroma barrier properties of edible films: a review. *Trends in Food Science and Technology*, **8**, 228–237.

Miller, K.S., Upadhyaya, S.K. and Krochta, J.M. (1998) Permeability of D-limonene in whey protein films. *Journal of Food Science*, **63**, 244–247.

Mohagheghzadeh, A., Faridi, P., and Ghasemi, Y. (2010) Analysis of Mount Atlas mastic smoke: a potential food preservative. *Fitoterapia*, **81**, 577–580.

Moreno, S., Sana, A.M.O., Gaya M., Barni, M.V., Castro, O.A. and Van Baren, C. (2012) Rosemary compounds as nutraceutical health products. In: *Food Additive* (El-Samragy, Y., ed.). InTech, pp. 157–174.

Murugesan, R. and Orsat, V. (2012) Spray drying for the production of nutraceutical ingredients – a review. *Food and Bioprocess Technology*, **5**, 3–14.

Nakagawa, K. and Nagao, H. (2012) Microencapsulation of oil droplets using freezing-induced gelatin–acacia complex coacervation. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, **411**, 129–139.

Naknean, P. and Meenume, M. (2010) Factors affecting retention and release of flavour compounds in food carbohydrates. *International Food Research Journal*, **17**, 23–34.

Nedovic, V., Kalusevic, A., Manojlovic, V., Levic, S. and Bugarsk, B. (2011) An overview of encapsulation technologies for food applications. *Procedia Food Science*, **1**, 1806–1815.

Neetoo, H. and Chen, H. (2012) Application of high hydrostatic pressure technology for processing and preservation of foods. In: *Progress in Food Preservation* (Bhat, R., Alias, A.K. and Paliyath, G., eds). Wiley-Blackwell, pp. 247–276.

Nussinovitch, A. (2003) *Water Soluble Polymer Applications in Foods*. Blackwell Science, Oxford, England, pp. 29–69.

Omobuwajo, T.O. (2007) Food flavourings: principles of applications. In: *Handbook of Food Products Manufacturing* (Hui, Y.H., ed.). John Wiley & Sons, Inc., Hoboken, New Jersey, USA, pp. 117–130.

Onwulata, C.I. (2012) Encapsulation of new active ingredients. *Annual Review of Food Science and Technology*, **3**, 183–202.

Ouattara, B., Simard, R. E., Piette, G., Bégin, A. and Holley, R.A. (2000) Inhibition of surface spoilage bacteria in processed meats by application of antimicrobial films prepared with chitosan. *International Journal of Food Microbiology*, **62**, 139–148.

Ozcalik, O. and Tihminlioglu, F. (2012) Barrier properties of corn zein nanocomposite coated polypropylene films for food packaging applications. *Journal of Food Engineering*, **114**, 505–513.

Park, J.W., Testin, R.F., Park, H.J., Vergano, P.J. and Weller, C.L. (1994) Fatty acid concentration effect on tensile strength, elongation, and water vapor permeability of laminated edible films. *Journal of Food Science*, **59**, 916–919.

Park, S.Y., Yoo, S.S., Shim, J.H. and Chin, K.B. (2008) Physicochemical properties, and antioxidant and antimicrobial effects of garlic and onion powder in fresh pork belly and loin during refrigerated storage. *Journal of Food Science*, **73**, C577–C584.

Pavlath, A. and Orts, W. (2009) Edible films and coatings: why, what, and how? In: *Edible Films and Coatings for Food Applications* (Embuscado, M.E. and Huber, K.C., eds). Springer, New York, USA, pp. 1–24.

Pereira, R.N., Souza, B.W.S., Cerqueira, M.A., Teixeira, J.A. and Vicente, A.A. (2010) Effects of electric fields on protein unfolding and aggregation: influence on edible films formation. *Biomacromolecules*, **11**, 2912–2918.

Pérez-Gago, M.B. and Krochta, J.M. (2000) Drying temperature effect on water vapor permeability and mechanical properties of whey protein–lipid emulsion films. *Journal of Agricultural and Food Chemistry*, **48**, 2687–2692.

Pérez-Gago, M.B. and Krochta, J.M. (2005) Emulsion and bi-layer edible films. In: *Innovations in Food Packaging* (Han, J.H., ed.). Elsevier Academic, Oxford, England, pp. 384–402.

Perkins-Veazie, P. and Collins, J.K. (2001) Contributions of nonvolatile phytochemicals to nutrition and flavor. *Horttechnology*, **11**, 539–546.

Phan, T. D., Debeaufort, F., Peroval, C., Despre, D., Courthaudon, J.L. and Voilley, A. (2002) Arabinoxylan–lipid-based edible films. 3. Influence of drying temperature on emulsion stability and on film structure and functional properties. *Journal of Agricultural and Food Chemistry*, **50**, 2423–2428.

Pokorny, J. and Trojáková, L. (2001) The use of natural antioxidants in food products of plant origin. In: *Antioxidants in Food: Practical Applications* (Pokorny, J., Yanishlieva, N. and Gordon, M., eds). Woodhead Publishing Ltd, CRC Press, pp. 355–372.

Prakash, B., Singh, P., Kedia, A. and Dubey, N.K (2012) Assessment of some essential oils as food preservatives based on antifungal, antiaflatoxin, antioxidant activities and in vivo efficacy in food system. *Food Research International*, **49**, 201–208.

Pranoto, Y., Rakshit, S.K. and Salokhe, V.M. (2005) Enhancing antimicrobial activity of chitosan films by incorporating garlic oil, potassium sorbate and nisin. *Food Science and Technology*, **38**(8), 859–865.

Pranoto, Y., Salokhe, V.M. and Rakshit, S.K. (2005) Physical and antibacterial properties of alginate-based edible film incorporated with garlic oil. *Food Research International*, **38**, 267–272.

Prata, A.S., Zanin, M.H.A., Ré, M.I. and Grosso, C.R.F. (2008) Release properties of chemical and enzymatic crosslinked gelatin–gum Arabic microparticles containing a

fluorescent probe plus vetiver essential oil. *Colloids and Surfaces B: Biointerfaces*, **67**, 171–178.

Pundir, R. K., Pranay, J. and Chetan, S. (2010) Antimicrobial activity of ethanolic extracts of *Syzygium aromaticum* and *Allium sativum* against food associated bacteria and fungi. *Ethnobotanical Leaflets*, **14**, 1–10.

Qi, H., Hu, W., Jiang, A., Tian, M. and Li, Y. (2011) Extending shelf-life of fresh-cut 'Fuji' apples with chitosan-coatings. *Innovative Food Science and Emerging Technologies*, **12**, 62–66.

Quezada-Gallo, J.A. (1999) Influence of structure and composition of macromolecular networks on volatile molecules (water and aromas) transfers. Application to edible packaging and plastics. PhD Dissertation, University of Dijon, France.

Ramos, O.L., Pereira, J.O., Silva, S.I., Fernandes, J.C., Franco, M.I., Lopes-da-Silva, J.A., Pintado, M.E. and Malcata, F.X. (2012) Evaluation of antimicrobial edible coatings from a whey protein isolate base to improve the shelf life of cheese. *Journal of Dairy Science*, **95**, 6282–6292.

Ramos-García, M., Bosquez-Molina, E., Hernández-Romano J., Zavala-Padilla G., Terrés-Rojas E., Alia-Tejacal, I., Barrera-Necha L., Hernández-López, M. and Bautista-Baños, S. (2012) Use of chitosan-based edible coatings in combination with other natural compounds, to control *Rhizopus stolonifer* and *Escherichia coli* DH5α in fresh tomatoes. *Crop Protection*, **38**, 1–6.

Ratanatriwong, P., Barringer, S.A. and Delwiche, J. (2003) Sensory preference, coating evenness, dustiness, and transfer efficiency of electrostatically coated potato chips. *Journal of Food Science*, **68**(4), 1542–1547.

Regnier, T., Combrinck, S. and Du Plooy, W. (2012) Essential oils and other plant extracts as food preservatives. In: *Progress in Food Preservation* (Bhat, R., Alias, A.K. and Paliyath, G., eds), Wiley-Blackwell, pp. 539–579.

Regulation (EC) No. 1331/2008 (2008) Establishing a common authorisation procedure for food additives, food enzymes and food flavourings. *Official Journal of the European Union*, 31 December 2008. http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2008:354:0001:0006:EN:PDF. Accessed November 2012.

Reineccius, G.A. (1989) Flavor encapsulation. *Food Reviews International*, **5**(2), 147–176.

Reineccius, G.A. (2009) Edible films and coatings for flavor encapsulation. In: *Edible Films and Coatings for Food Applications* (Embuscado, M.E. and Huber, K.C., eds). Springer Science Business Media, New York, USA, pp. 269–294.

Ribeiro, C., Vicente, A.A., Teixeira, J.A. and Miranda, C. (2007) Optimization of edible coating composition to retard strawberry fruit senescence. *Postharvest Biology and Technology*, **44**, 63–70.

Rijk, R. (2007) Chemical migration from active and intelligent packaging materials into food. In: *Chemical Migration and Food Contact Materials* (Barnes, K.A., Sinclair, C.R. and Watson, D.H., eds). Woodhead Publishing Limited, pp. 371–391.

Risch, S.J. and Reineccius, G.A. (1988) Effect of emulsion size on flavor retention and shelf-stability of spray dried orange oil. In: *Encapsulation and Controlled Release of Food Ingredients* (Reineccius, G.A. and Risch, S.J., eds). American Chemical Society, Washington, DC, USA, p. 67.

Rodriguez-Turienzo, L., Cobos, A. and Diaz, O. (2012) Effects of edible coatings based on ultrasound-treated whey proteins in quality attributes of frozen Atlantic salmon (*Salmo salar*). *Innovative Food Science and Emerging Technologies*, **14**, 92–98.

Rojas-Graü, M.A., Avena-Bustillos, R.J., Olsen, C., Friedman, M., Henika, P.R., Martín-Belloso, O., Pan, Z. and McHugh, T.H. (2007a) Effects of plant essential oils and oil compounds on mechanical, barrier and antimicrobial properties of alginate–apple puree edible films. *Journal of Food Engineering*, **81**, 634–641.

Rojas-Graü, M.A., Raybaudi-Massilia, R.M., Soliva-Fortuny, R.S., Avena-Bustillos, R.J., McHugh, T.H. and Martín-Belloso, O. (2007b) Apple puree–alginate edible coating as carrier of antimicrobial agents to prolong shelf-life of fresh-cut apples. *Postharvest Biology and Technology*, **45**, 254–264.

Roos, Y. and Karel, M. (1991) Plasticizing effect of water of thermal behaviour and crystallization of amorphous food models. *Journal of Food Science*, **56**, 38–43.

Ruiz-Navajas, Y., Viuda-Martos, M., Sendra, E., Perez-Alvarez, J.A. and Fernández-López, J. (2013) *In vitro* antibacterial and antioxidant properties of chitosan edible films incorporated with *Thymus moroderi* or *Thymus piperella* essential oils. *Food Control*, **30**, 386–392.

Salzer, U.-J. (2007) Legislation/toxicology. In: *Flavourings Production, Composition, Applications, Regulations*, 2nd edn (Ziegler, H., ed.). Wiley-VCH Verlag GmbH & Co., KGaA, Weinheim, Germany.

Sánchez-González, L., Vargas, M., Gonzalez-Martinez, C., Chiralt, A. and Chafer, M. (2009) Characterization of edible films based on hydroxypropylmethylcellulose and tea tree essential oil. *Food Hydrocolloids*, **23**, 2102–2109.

Sánchez-González, L., Chafer, M., Chiralt, A. and Gonzalez-Martinez, C. (2010a) Physical properties of edible chitosan films containing bergamot essential oil and their inhibitory action on *Penicillium italicum*. *Carbohydrate Polymers*, **82**, 277–283.

Sánchez-González, L., Gonzalez-Martinez, C., Chiralt, A. and Chafer, M. (2010b) Physical and antimicrobial properties of chitosan–tea tree essential oil composite films. *Journal of Food Engineering*, **98**, 443–452.

Santas, J., Almajano, M.P. and Carbó, R. (2010) Antimicrobial and antioxidant activity of crude onion (*Allium cepa* L.) extracts. *International Journal of Food Science and Technology*, **45**, 403–409.

Sarikurkcu, C., Sabih Ozer, M., Eskici, M., Tepe, B., Can, S. and Mete, E. (2010) Essential oil composition and antioxidant activity of *Thymus longicaulis* C. Presl subsp. longicaulis var. longicaulis. *Food and Chemical Toxicology*, **48**, 1801–1805.

Sarkar, S., Gupta, S., Variyar, P.S., Sharma, A. and Singhal, R.S. (2012) Irradiation depolymerized guar gum as partial replacement of gum Arabic for microencapsulation of mint oil. *Carbohydrate Polymers*, **90**, 1685–1694.

Sasidharan, I. and Menon, A.N. (2010) Comparative chemical composition and antimicrobial activity fresh and dry ginger oils (Zingiber Officinale Roscoe). *International Journal of Current Pharmaceutical Research*, **2**, 40–43.

Saucedo-Pompa, S., Rojas-Molina, R., Aguilera-Carbó, A.F., Saenz-Galindo, A., de La Garza H., Jasso-Cantú, D. and Aguilar, C.N. (2009) Edible film based on candelilla wax to improve the shelf life and quality of avocado. *Food Research International*, **42**, 511–515.

Sayanjali, S., Ghanbarzadeh, B. and Ghiassifar, S. (2011) Evaluation of antimicrobial and physical properties of edible film based on carboxymethyl cellulose containing potassium sorbate on some mycotoxigenic *Aspergillus* species in fresh pistachios. *LWT-Food Science and Technology*, **44**, 1133–1138.

Shaikh, J., Bhosale, R. and Singhal, R. (2006) Microencapsulation of black pepper oleoresin. *Food Chemistry*, **94**, 105–110.

Shimoni, E. (2009) Nanotechnology for foods: delivery systems. In: *Global Issues in Food Science and Technology* (Barbosa-Canovas, G.V., Mortimer, A., Lineback, D., Spiess, W., Buckle, K. and Colonna, P., eds). Elsevier Science, pp. 411–424.

Shinde, U. and Nagarsenker, M. (2011) Microencapsulation of eugenol by gelatin sodium alginate complex coacervation. *Indian Journal of Pharmaceutical Sciences*, **73**, 311–315.

Shukla, R., Singh, P., Prakash, B. and Dubey, N.K. (2012) Antifungal, aflatoxin inhibition and antioxidant activity of *Callistemon lanceolatus* (Sm.) sweet essential oil and its major component 1,8-cineole against fungal isolates from chickpea seeds. *Food Control*, **25**, 27–33.

Smith, R.L., Waddell, W.J., Cohen, S.M., Feron, V.J., Marnett, L.J., Portoghese, P.S., Rietjens, I.M.C.M., Adams, T.B., Gavin, C.L., McGowen, M.M., Taylor, S.V. and Williams, M.C. (2009) GRAS flavoring substances 24. *Food Technology*, **46–105**.

Soher E.A., Aziza A.S., Moshira E.S, Khaled F.E. and Bassem A.S (2011) Chemical profile, antioxidant, antifungal and antiaflatoxigenic activity of parsley and ginger volatile and non-volatile extracts. *Journal of Biologically Active Products from Nature*, **1**, 81–96.

Soković, M., Glamočlija, J., Marin, P.D., Brkić, D. and Van Griensven, L.J.L.D. (2010) Antibacterial effects of the essential oils of commonly consumed medicinal herbs using an *in vitro* model. *Molecules*, **15**, 7532–7546.

Soottitantawat, A., Yoshii, H., Furuta, T., Ohkawara, M. and Linko, P. (2003) Microencapsulation by spray drying: influence of emulsion size on the retention of volatile compounds. *Journal of Food Science*, **68**, 2256–2262.

Soradech, S., Nunthanid, J., Limmatvapirat, S. and Luangtana-anan, M. (2012) An approach for the enhancement of the mechanical properties and film coating efficiency of shellac by the formation of composite films based on shellac and gelatin. *Journal of Food Engineering*, **108**, 94–102.

Sothornvit, R. and Krochta, J.M. (2000) Plasticizer effect on oxygen permeability of beta–lactoglobulin films. *Journal of Agricultural and Food Chemistry*, **48**, 6298–6302.

Sothornvit, R. and Krochta, J.M. (2001) Plasticizer effect on mechanical properties of beta–lactoglobulin films. *Journal of Food Engineering*, **50**, 149–155.

Souza, B.W.S., Cerqueira, M.A., Casariego, A., Lima, A.M., Teixeira, J.A. and Vicente, A.A. (2009) Effect of moderate electric fields in the permeation properties of chitosan coatings. *Food Hydrocolloids*, **23**, 2110–2115.

Stojanovic, R., Belscak-Cvitanovic, A., Manojlovic, V., Komes, D., Nedovic, V. and Bugarski, B. (2011) Encapsulation of thyme (*Thymus serpyllum* L.) aqueous extract in calcium alginate beads. *Journal of the Science of Food and Agriculture*, **92**(3), 685–696.

Suarez, R.B., Campanone, L.A., García, M.A. and Zaritzky, N.E. (2008) Comparison of the deep frying process in coated and uncoated dough systems. *Journal of Food Engineering*, **84**, 383–393.

Sun, C., Gunasekaran, S. and Richards, M.P. (2007) Effect of xanthan gum on physicochemical properties of whey protein isolate stabilized oil-in-water emulsions. *Food Hydrocolloids*, **21**, 555–564.

Sun-Waterhouse, D. and Wadhwa, S.S. (2012) Industry-relevant approaches for minimising the bitterness of bioactive compounds in functional foods: a review. *Food and Bioprocess Technology*, **3**, 607–627.

Tassou, C.C. and Nychas, G.J.E. (1995) Antimicrobial activity of the essential oil of mastic gum (*Pistacia lentiscus* var. chia) on Gram positive and Gram negative bacteria in broth and in Model Food System. *International Biodeterioration and Biodegradation*, **36**, 411–420.

Tassou, C., Koutsoumanis, K. and Nychas, G.J.E. (2000) Inhibition of *Salmonella enteritidis* and *Staphylococcus aureus* in nutrient broth by mint essential oil. *Food Research International*, **33**, 273–280.

Taylor, A.J. (1998) Physical chemistry of flavour. *International Journal of Food Science and Technology*, **33**, 53–62.

Thies, C. (2007) Microencapsulation of flavors by complex coacervation. In: *Encapsulation and Controlled Release Technologies in Food Systems* (Lakkis, J.M., ed.). Blackwell Publishing, Ames, Iowa, USA, pp. 149–170.

Tian, J., Huang, B., Luo, X., Zeng, H., Ban, X., He, J. and Wang, J. (2012) The control of *Aspergillus flavus* with *Cinnamomum jensenianum* Hand.-Mazz essential oil and its potential use as a food preservative. *Food Chemistry*, **130**, 520–527.

Turgis, M., Vu, K.D., Dupont, C. and Lacroix, M. (2012) Combined antimicrobial effect of essential oils and bacteriocins against foodborne pathogens and food spoilage bacteria. *Food Research International*, **48**, 696–702.

Ustunol, Z. (2009) Edible films and coatings for meat and poultry. In: *Edible Films and Coatings for Food Applications* (Embuscado, M.E. and Huber, K.C., eds). Springer, New York, USA, pp. 245–268.

Vicente, A.A., Cerqueira, M.A., Hilliou, L. and Rocha, C. (2011) Protein-based resins for food packaging. In: *Multifunctional and Nanoreinforced Polymers for Food Packaging* (Lagaron, J.M., ed.). Woodhead Publishing Limited, Cambridge, England, pp. 610–648.

Vodnar, D. (2012) Inhibition of *Listeria* monocytogenes ATCC 19115 on ham steak by tea bioactive compounds incorporated into chitosan-coated plastic films. *Chemistry Central Journal*, **6**, 74.

Weinbreck, F., Nieuwenhuijse, H., Robijn, G.W., and De Kruif, C.G. (2004) Complexation of whey proteins with carrageenan. *Journal of Agricultural and Food Chemistry*, **52**, 3550–3555.

Wu, Y., Weller, C.L., Hamouz, F., Cuppett, S.L. and Schnepf, M. (2002) Development and application of multicomponent edible coatings and films: a review. *Advances in Food and Nutrition Research*, **44**, 347–394.

Xie, Y.-L., Zhou, H.-M. and Zhang, Z.-R. (2007) Effect of relative humidity on retention and stability of vitamin A microencapsulated by spray drying. *Journal of Food Biochemistry*, **31**, 68–80.

Xing, F., Cheng, G., Yang, B. and Ma, L. (2004) Microencapsulation of capsaicin by the complex coacervation of gelatin, acacia and tannins. *Journal of Applied Polymer Science*, **91**(4), 2669–2675.

Yanishlieva, N.V., Marinova, E. and Pokorný, J. (2006) Natural antioxidants from herbs and spices. *European Journal of Lipid Science and Technology*, **108**, 776–793.

Ye, C.-H., Dai, D.-H. and Hu, W.-L. (2013) Antimicrobial and antioxidant activities of the essential oil from onion (*Allium cepa* L.). *Food Control*, **30**, 48–53.

Yeo, Y., Bellas, E., Firestone, W., Langer, R. and Kohane, D.S. (2005) Complex coacervates for thermally sensitive controlled release of flavor compounds. *Journal of Agricultural and Food Chemistry*, **53**(19), 7518–7525.

Yilmaz, F. and Dagdemir, E. (2012) The effects of beeswax coating on quality of Kashar cheese during ripening. *International Journal of Food Science and Technology*, **47**, 2582–2589.

Zahedi, Y., Ghanbarzadeh, B. and Sedaghat, N. (2010) Physical properties of edible emulsified films based on pistachio globulin protein and fatty acids. *Journal of Food Engineering*, **100**, 102–108.

Zaritzky, N. (2011) Edible coatings to improve food quality and safety. In: *Food Engineering Interfaces* (Aguilera, J., Barbosa-Cánovas G., Simpson, R., Welti-Chanes, J. and Bermúdez-Aguirre, D., eds), Springer, New York, pp. 631–659.

Zhang, Y. and Han, J.H. (2006) Mechanical and thermal characteristics of pea starch films plasticized with monosaccharides and polyols. *Journal of Food Science*, **71**(2), 109–118.

Zhang, C., Gao, D., Ma, Y. and Zhao X. (2013) Effect of gelatin addition on properties of pullulan films. *Journal of Food Science*, **78**, C805–C810.

Zhao, Y. and McDaniel, M. (2005) Sensory quality of foods associated with edible film and coating systems and shelf-life extension. In: *Innovations in Food Packaging*. Han, J.H. (Ed.). Elsevier Academic Press, San Diego, CA. pp. 434–453.

Zivanovic, S., Chi, S. and Draughon, A.F. (2005) Antimicrobial activity of chitosan films enriched with essential oils. *Journal of Food Science*, **70**(1), 1145–1151.

13

Instantization and Agglomeration of Foods

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13.1 Introduction

The concept of instantization has arisen due to the primary need of providing convenience to consumers by modifying the functional behaviour of raw materials as well as products. The convenience may be quantified in terms of the time of preparation while qualitative improvements are possible in terms of ease in preparation. Further, easy flowability and/or dispersability are the other desirable characteristics that make the finished product more attractive and convenient for the end users. In addition, social factors such as the increase in the number of nucleus families, change in lifestyles and more emphasis on professional activities have reduced the time available for preparation of meals and other associated activities. An increase in the capacity to purchase processed foods and the desire to have more leisure time has led to the increased demand for convenience and instant foods.

The word 'instant food' was earlier used for different types of pre-mixes, but these are truly not instant foods as they may require preparation time of more than 5 min. Such products can at best be called convenience foods. Examples include several cake mixes and *dosa/idli* (traditional Indian foods based on rice and blackgram) mixes.

There is no universally accepted definition for instant foods. The word 'instant food' refers to a ready-to-use or consumable form. These can be prepared from dry mixes, powders, liquids, extracts, concentrates and emulsions

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using different formulations (Villagran *et al.*, 2002). Their preparation may require process technologies such as drying (convection/spray/freeze drying), cryotechniques, expansion process heating or extrusion (Youssef, 1990).

The first attempt to establish an international definition was made at the International Dairy Federation session in Moscow in 1968. Skim milk powder is considered instant if, under standard stirring conditions, all lumps disappear within 15 s (Jensen, 1973). The definition that can be generated now for instant food is that it is a kind of food that helps reduce the time of preparation/cooking and drudgery, which ultimately provides convenience to consumers. In a quantitative sense, an instant food does not require more than 5 min of time between the start of preparation and the actual consumption by the end user. A quick cooking noodle falls into this category. Processed food requiring a preparation time of less than 1 min (e.g. instant tea powder) may be categorized as an ultra-instant food.

Instantization is thus a process of developing instant properties (like easy dissolution, rehydration, etc.) in a food product. Agglomeration is one of the processing methods to make the product instant (Youssef, 1990). Many instant products available on the shelves are made from various raw materials and other ingredients in order to develop special features, like easy processing/cooking, functional benefits and amenability for fortification. The following sections discuss these instant products and the methods of their preparation.

Agglomeration is a physical phenomenon and can be described as the sticking of particulate solids, which is caused by short-range physical or chemical forces among the particles. This phenomenon is enhanced by selecting appropriate processing conditions or binders and substances that adhere chemically or physically to form bridges between particles. The main purpose of particle size enlargement by agglomeration is to control certain physical properties of food powders, such as density, flowability, to improve dispersion and dissolution characteristics and to reduce the tendency of caking and dust formation (Mukherjee and Bhattacharya, 2006).

13.2 Applications of the technology/process

Instant foods may be grouped into seven categories depending on the type of the major raw material used, namely foods based on cereals (Table 13.1), legumes (Table 13.2), vegetables (Table 13.3), meat, fish and poultry (Table 13.4), beverages (Table 13.5), milk (Table 13.6) and miscellaneous products (Table 13.7).

13.2.1 Cereal-based instant products

The raw and processed flours from rice, corn, wheat and their starches, semolina and even the whole grains are used as the basic raw material for

Table 13.1 Important cereal-based instant products

Name of product	Major raw material(s)	Processing technology employed	References
Instant pudding mix	Starch, sugar	Mixing of all raw materials	Katt, Moore and Eastman (1986)
Dried instant food	Starch, pregelatinized starch, modified ungelatinized starch	Agglomeration and mixing with dried food ingredients	Bohrmann, Bezner and Sidler (1979)
Flavoured flaky instant food	Starch, wheat flour, wheat semolina	Roller, fluidized bed and final drying for flavour adhesion	Zameitat (1976)
Instant soakable rice	Rice grain	Soaking–steaming–soaking–drying–puffing	Lee & Wissgott (2001)
Instant rice flake	Rice grain	Soaking–cooking–drying–flaking–final drying	Eu-Chin (2000)
Instant modified flour	Wheat, rye, corn, potato, tapioca	Conditioning–extrusion cooking–pelleting–dry milling–moistening–mixing–agglomeration	Bruemmer (2003)
Instant flaked/beaten rice	Rough rice	Hydration–cooking–flaking–drying	Mujoo and Ali (2000)

developing commercially important cereal-based instant foods (Table 13.1). The product, such as instant flaked/beaten rice, is a convenience food requiring only soaking for a short duration at room temperature prior to consumption. As an example, the development of instant (soakable) rice is shown in Figure 13.1 where rice grain is subjected to two cycles of soaking and steaming followed by partial drying and puffing (Lee and Wissgott, 2001). In a starch-rich raw material, instantization is achieved by gelatinization of starch to ensure a low processing time at the consumers' end.

13.2.2 Pulse-based products

Apart from cereals, the starch-protein rich pulses are frequently used for developing instant foods. The hard texture of several pulse/legume-based raw materials are tackled by subjecting them to hydrothermal treatments for gelatinization of starch, inactivation of antinutritional factors and denaturation of proteins (Table 13.2). As the raw legumes (whole or split) conventionally take a long time to cook, there is a need for developing instant products to offer convenience to the end users. The flowchart for the development of instant glass noodles is shown in Figure 13.2. Steaming causes gelatinization of

Table 13.2 Legume-based instant products

Name of product	Major raw materials	Processing technology employed	References
Instant whole meal protein extrudates	Whole wheat flour, malted whole grain meals, apple fruit powder	Extrusion–cooked	Mueller (1986)
Dried beans/ redgram as instant food	Broad beans (*Vicia faba*), adzuki (*Phaseolus angularis*)	Boiling in water–enzymatic and acid treatment–dried with superheated steam	Murata, Shimizu and Kokeguchi (1982)
	Raw beans, lactose/sucrose	Boiling in water–enzymatic treatment–sugar coating	Hase, Yamauchi and Handa (1980)
	Red gram	Cooking–fluidized bed drying	Bhuibhar *et al.* (1991)
Instant glass noodles	Mung bean, potato starch	Extrusion–steaming-blanching–chilling–drying to <15% moisture content	Lian and Toh (1998)
Cereal–legume based instant mix (*dhokla*)	Rice, Bengal gram, black gram	Grinding–sieving–blending	Mahajan and Chattopadhyay (2000)

Table 13.3 Vegetable-based instant products

Name of product	Major raw materials	Processing technology employed	References
Instant food product	Potato puree powder/meal, powdered fat, modified starch	Mixing–heating–stirring/blending	Antwi-Afriyie (2005)
Vegetable-based instant food	Vegetables, wheat flour, vinegar, reducing sugars	Heating–drying	Aleksandrovich (2001)
Instant puree powder	Berries, vegetables, fruits	Mixing–freezing–size reduction–agglomeration/ granulation–drying	Strommen, Eikevik and Alves-Filho (2001)

starch while chilling at the end of processing provides the necessary nominal retrogradation to obtain a firm texture and appearance (Lian and Toh, 1998).

13.2.3 Vegetable-based foods

Vegetable-based instant food products include instant soup/puree/ concentrate, with puree powder being less in number (Table 13.3). Thermal

Table 13.4 Instant products based on meat, fish and poultry

Name of product	Major raw materials	Processing technology employed	References
Dry instant food	Vegetables, refined/unrefined products of meat and fish	Heating – mixing – freezing – drying	Nove (2007)
Shark fin-based instant food	Harusame, arrow root flour, jelly fish, starch	Injection moulding	Yoshitoshi (2006)
Meat-based instant food	Beef, rabbit and poultry meat, fish and sea food	Heating – crushing – forming – drying	Aleksandrovich (2002)
Instant gelatin	Gelatin obtained from animal sources	Soaking – agglomeration – drying in fluidized bed	Maini and Maini (2011)

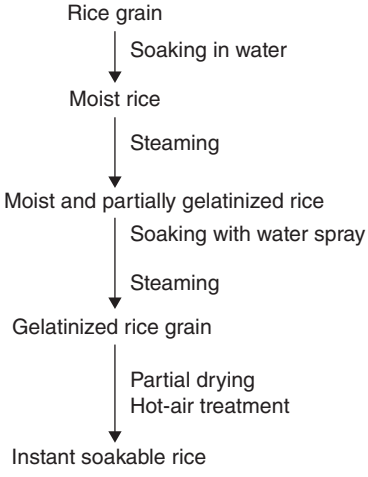

Figure 13.1 Flowchart for the preparation of instant soakable rice

treatments, mixing, drying and agglomeration are the common processing steps that induce instant characteristics in the finished product. Another product *fufu*, a delicacy from Ghana, is prepared by mixing potato powder along with other ingredients followed by heating and stirring/blending to develop instant *fufu* (Antwi-Afriyie, 2005).

13.2.4 Instant products of animal origin

Meat, fish and poultry-based instant products are shown in Table 13.4. Fish, egg and meat from different animal sources are subjected to thermal processing to obtain instant products. A patented literature by Nove (2007) claims to

Table 13.5 Instant beverage products

Name of product	Major raw materials	Processing technology employed	References
Instant food product/beverage powder	Sugar, tea, coffee/fruit extracts, cocoa powder	Moistening–moulding–drying	Groote, Rainer and Tomas (2006)
	Corn syrup solids, poly fatty acids, sucrose	Mixing–agglomeration–sizing–drying	Butterbaugh and Sargent (2002)
	Ground coffee beans/tea leaves, sweetener, sugars, dried milk products, gelatin	Mixing–compaction to form tablets	Kenke and Walkowiak (2000)
	Coconut milk powder	Spraying–drying	Steiger (2006)
	Etherified oils, terpenes, vegetable oils	Mixing–homogenization–spray drying	Kumlehn *et al.* (2004)
	Soy protein	Mixing	Gallen (2002)
	Protein source, nondairy creamer, sodium caseinate, corn syrup solids, vegetable gum, carbohydrate source	Dry mixing of all raw materials to make a nutritional drink	Schechter (2000)
	Cappuccino powder, cocoa powder mixes, tea dust	Agglomeration–fluidized bed drying	Waskow, Ruempler and Jacob (2005)
	Water, emulsifier, stabilizer, oils, terpenes, vegetables oils	Homogenization–spray drying	Kumlehn *et al.* (2004)

Instant tea	Tea leaves, pectin	Extraction–spray drying	Gurkin *et al.* (1972)
	Green/black tea, ginseng, chrysanthemum, citric acid, sugar	Extraction filtration–concentration–mixing–spray drying to prepare nutritious tea	Liying (2009)
	Tea leaves	Extraction–deleafing–concentration–addition of anionic colloid decreaming–final concentration–drying to prepare cold water soluble tea concentrate	George and Saksena (2002)
Instant coffee powder	Coffee powder, creamer, whitener, deionized water	Spray drying	Kessler *et al.* (2010)
	Coffee powder, sweetener, maple syrup flavour	Dry mixing/wet mixing–freeze drying	Fairhurst, Labbe and Ortega (2002)
	Gluconolactone, alkali metal carbonate/ bicarbonate, sweetener	Mixing–granulation–drying to make coffee creamer	Zeller (1998)
Instant chocolate drinks	Cocoa solids, stabilizing agent, microcrystalline cellulose, starch, maltodextrin, nonfat dry milk	Fluidized bed drying/fluidized spray drying	Tuason and McGinley (1990)
Instant powder for alcoholic beverage	Monoethylcarbonate, citric acid, flavours, sweeteners	Spray drying	Sterzel (2001)

Table 13.6 Milk-based instant food products

Name of product	Major raw materials	Processing technology employed	References
Instant milk powders	Whole milk, skim milk	Re-wet agglomeration	Upadhyay, Vyas and Pandya (1982)
Instant dried whole milk	Whole milk, lecithin, butter oil	Spray drying–coating with lecithin	Pisecky and Westergaard (1972)
Instant whole milk powder	Whole milk	Fluidized bed agglomeration–coating	Baldwin and Sanderson (1972)

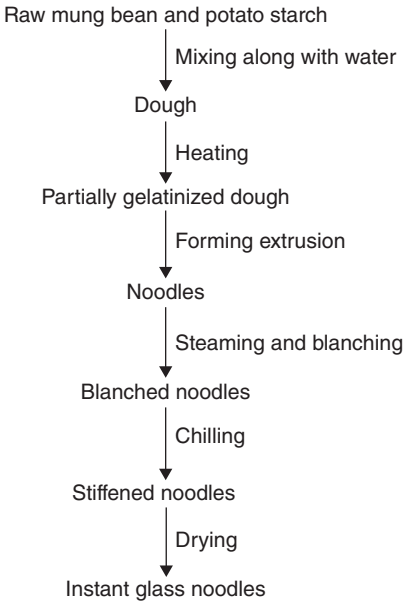

Figure 13.2 Flowchart for the preparation of instant glass noodles

have developed an instant product for infants that is rehydratable in less than 3 min upon addition of hot water (Figure 13.3). The mixture of ingredients is cooled to a temperature between –3 and –5 °C followed by two stages of drying. A fluidized bed dryer has been employed and the finished product has a moisture content of less than 8%.

Table 13.7 Miscellaneous instant products

Name of product	Major raw materials	Processing technology employed	Reference
Nutrient enriched instant food	Vegetables, fruit/meat/milk and cereals, spices, fibre, minerals and vitamins	Drying–size reduction	Denis and Petra (2006)
Instant Egyptian food	Skim milk powder, rice, wheat, beans	Autoclaving–dehydration	Youssef (1990)
Instantized powder drinks	Skim milk and cocoa powder	Re-wet agglomeration–instantisation	Jensen (1973)
Instant food mix	Vegetable and animal products, wheat flour, tartaric acid, vinegar, disaccharides, surfactants	Mixing–heating	Ryk (2001)
Instant food and feed	Milk, whey, egg, animal fat, wheat/corn starch, vegetable oil, lactose, maltodextrin, coffee/cocoa beverages, fruit juice/concentrate, yeast extract	Re-wetting of dried powder–agglomeration–drying in fluidized bed	Quandt *et al.* (2010)
Readily dispersible sugar product	Sugar syrup with high solid content	Crystallization	Chen *et al.* (1982)

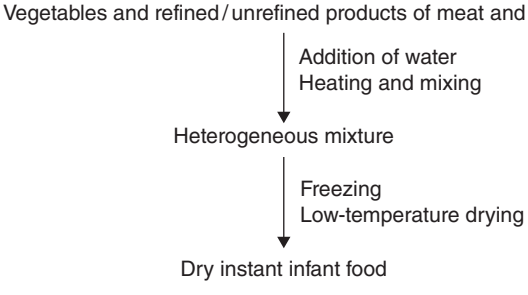

Figure 13.3 Processing steps for the development of dry instant infant food

13.2.5 Instant beverage products

Several instant beverage products have been developed and examples include instant powder made from tea, coffee, coconut milk and cocoa. In addition, instant coffee-milk, decaffeinated instant green tea as a nutritious (health) drink, coffee creamer and chocolate drink have been reported (Table 13.5). A large number of ingredients including coffee beans, tea leaves, cocoa powder, emulsifier like lecithin, hydrocolloids like alginate and pectin, protein sources (soy protein, caseinate, gelatin), flavouring and colouring agents, and different sugars are used to develop these products. Agglomeration/drying such as conventional drying, fluidized bed drying, spray drying and freeze drying are frequently employed to develop the instantized beverage powders and products. In all these products, the emphasis is on the quick dispersability of the developed product. Kenke and Walkowaik (2000) have developed an instant beverage tablet using coffee beans or tea leaves wherein the ingredients are finally compacted to offer the shape of a tablet for providing convenience to consumers.

13.2.6 Instant dairy products

Instant milk powder is possibly the oldest technology among the instant food products. Instant milk-based powders can be prepared from whole milk or skim milk with several additives such as sugar, cocoa powder, fruit juice/pulp, yoghurt, butter milk and lecithin added (Table 13.6). In the process shown in Figure 13.4 (Upadhyay, Vyas and Pandya, 1982), the wetting of skim milk

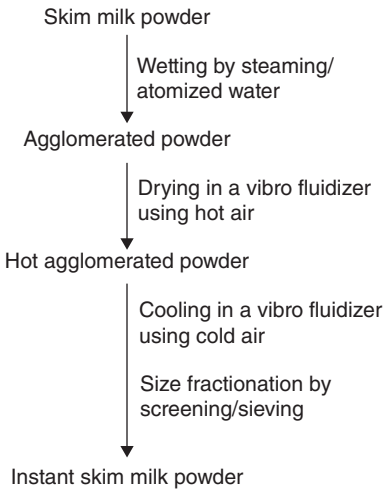

Figure 13.4 Flowchart for the preparation of instant skim milk powder

powder is done by steam/atomized water for creating sticky surface particles that induce the process of agglomeration. Later, these agglomerated particles are quickly dried and cooled to obtain instant skim milk powder. The product shows good wetting, sinking, dispersing and dissolving ability. Lecithin at a low concentration (0.2%) acts as a surface active agent to stabilize the fat globules in different milk-based products (Baldwin and Sanderson, 1972), leading to improvement in the wetting properties.

13.2.7 Miscellaneous instant products

Instant food products, not grouped into the categories, are mentioned in Table 13.7. These products include instant edible gel, enriched instant food and sugar-based products. Generally, processes like mixing, drying, wet heating and agglomeration are used to make these instant products. Youssef (1990) has reported the instantization and evaluation of some traditional Egyptian foods such as kishk, bellila and rice-milk. The processing steps for the production of rice-milk includes the cooking of rice grains in water followed by drying, size reduction and mixing with skim milk powder and sugar to obtain instant rice-milk (Figure 13.5). The calculated protein efficiency ratio (C-PER) for this product has been reported to be 2.4 while the *in vitro* protein digestibility is 89.3.

It appears that although many instant products have been developed in different countries, the technologies are mostly protected by patents, indicating their commercial importance and applications. However, the available information indicates that a wide variety of raw materials is used while wet heating, drying and agglomeration are the commonly employed processing technologies.

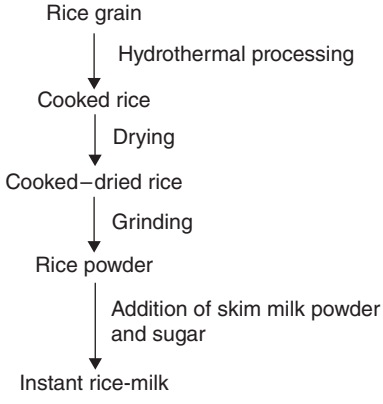

Figure 13.5 Process flowchart for the development of the traditional Egyptian instant rice-milk

13.3 Process technology

Common processing technologies and machines employed in the production of instant foods are summarized in Table 13.8. Frequently, agglomeration and drying techniques are an integral part of such process operations and product development.

13.3.1 Agglomeration

Agglomeration is one of the most commonly used process technologies to produce several instant food products (Table 13.9). It is also known as the size enlargement process to decrease density (though in some cases density may increase due to compaction) where the size of the agglomerates usually ranges between 0.2 and 2.0 mm. The particle size, shape (Figure 13.6), colour and strength of agglomerates are of main concern. Dairy powders, convenience foods, instant beverages, confectionery products and cereals are some of the products obtained from different agglomeration processes (Palzer, 2007). Agglomeration can be carried out by a wide variety of processes such as fluidized bed agglomeration, pressure agglomeration (compaction), granulation (dry and wet) and steam jet agglomeration.

In fluidized bed agglomeration, the upward flow of air in a fluidization chamber occurs in the batch and continuous mode of operation. The main purpose here is to provide uniform process conditions to the individual particles of the raw material to be processed. Usually instant milk and beverage powders are produced using this technology (Peitsch, 2002).

The pressure agglomeration process is carried out by applying external pressure on to the raw material. Agglomerated products, which are produced using a binder, are of low/medium strength. Therefore, to obtain stronger bonds subsequent drying/heating may be necessary. Piston presses, roller presses, isostatic presses and extrusion machinery are the most commonly used equipments. The main advantages are that the process is independent of the particle size of the raw material used and it is best suited for small production runs. Based on the extent of the pressure applied it can be subdivided as high-, medium- and low-pressure processes (Peitsch, 1991). The size enlargement process where no binder is used and the material is subjected to higher operating pressures is known as binderless pressure agglomeration to form tablets/briquettes. The medium- and low-pressure processes where the binder is often used is called pelleting (Peitsch, 2002).

Agglomeration of powdery food materials can also be achieved by granulation. It mainly aims at the production of granules of some specific size and shape of the raw material. It works on the principle of a sieving mechanism where an additional blade is provided in a granulator for an external pressure application and efficient processing. Two types of granulation process are in

Table 13.8 Application of common machineries for the manufacture of instant foods

Unit operation	Machinery used	Application	Products	References
Drying	Spray dryer	Drying liquid or semi-liquid sample to powdery mass	Milk powder, ice cream mix, soup/puree powders	Gong *et al.* (2007)
	Conventional tray/through flow/rotary dryer	Formation of strong solid bridges between particles and bring down moisture content	Skim and whole milk powder, maltodextrins, soup powders	Palzer (2007)
	Freeze dryer	Lowering the moisture content of heat-sensitive material	Vegetables, meat, fish and other perishable commodities	Nove (2007)
	Drum dryer	Cooking and drying of easily flowable solutions/dispersions/pastes	Instant potatoes, baby cereal foods, corn syrup, starchy material	Baker (1997)
Agglomeration	Agglomerator	Formation of agglomerates	Beverage powders such as coffee/chicory powder, starches	Jensen (1975); Dhanalakshmi and Bhattacharya (2011)
	Fluidized bed granulator	Spraying and forming granules	Corn flour granules, beverage powders such as coffee and cocoa	Turchiuli *et al.* (2005)
	Compaction agglomerator	Formation of high-density agglomerates of variety of shapes	Corn flour pellets, corn starch	Yusof, Smith and Briscoe (2005); Mukherjee and Bhattacharya (2006)
	Steam jet agglomerator/ instantizer	Steaming for heating the powdery material, agglomerate formation and final drying	Sugar with coffee extract/cocoa powder, maltodextrin/ corn starch powder	Schuchmann (1995); Takeiti, Kieckbusch and Collares-Queiroz (2008)

(*continued overleaf*)

Table 13.8 (*continued*)

Unit operation	Machinery used	Application	Products	References
	Rotating disc agglomerator	Moistening, agglomerate formation and drying	Instant coffee powder/granule	Stoltze and Masters (1979)
	Vibro fluidizer	Obtaining powder of good instant properties	Instant milk powder, chocolate drink powder	Jensen (1975)
	Re-wet instantizer	Droplet agglomeration to obtain small agglomerates	Cocoa/sugar mixtures, baby food powder and beverage whiteners	Jensen (1973)
	Mixer	For uniform distribution of powdery solids, moisture and binder liquid	Mixing of solids/ powdery materials	Kuti (1978)
	Extrusion cooker	Gelatinization and for binding of particles together	Wheat/rice/ corn flours, instant tea, seasonings, animal feed	Mueller (1986)
	Autoclave	Hydrothermal treatment to starchy material	Instant soakable rice, agglomerated corn/rice starch	Lee and Wissgott (2001)
	Sieve	Obtaining uniform sized particles and formation of agglomerates/granules	Flours of rice, wheat, corn, black gram, Bengal gram	Mahajan and Chattopadhyay (2000)

Table 13.9 Classification of agglomerated products

Major raw material used	Major ingredient(s)	Product	Processing technology employed	References
Cereals	Wheat flour, protein rich meal	Free flowing wheat flour granules	Granulation in fluidized bed	Berizzi (2004)
	Rice/other cereals, herbs, spices, vegetables	Flavoured rice	Agglomeration using ultrasonic energy	Capodieci (2002)
	Corn starch, gum Arabic, maltodextrin	Model system (agglomerates corn starch)	Hydration – addition of binder – drying	Ghosal, Indira and Bhattacharya (2010)
	Native and pre-compacted corn starch powder	Agglomerated starch with improved flow property	Agglomeration by fluidization/ spray drying	Cunningham (2007)
	Hydrolysed and unhydrolysed gelatin, starch hydrolysate	Model system (agglomerated granules)	Top spray fluidized bed coating	Dewettinck *et al.* (1999)
	Rice flour	Compacted rice flour mass	Hydration – compaction agglomeration – drying	Mukherjee and Bhattacharya (2006)
	Pre-gelatinized starch, gum Arabic	Agglomerated and readily dispersible powder	Mixing – spray drying	Zhao and Bertrand (2004)
Legume	Soy milk, maltodextrin	Instant soy milk powder	Ultrafiltration – spray drying – fluidized bed agglomeration	Jinapong, Suphantharika and Jamnong (2008)
Vegetables	Onion, sweet corn/ potato, green bean, tomato, red beet, asparagus, spinach, peppers	Instant free flowing vegetable powders	Mixing – drum drying	Cremer *et al.* (2008)

(*continued overleaf*)

Table 13.9 (*continued*)

Major raw material used	Major ingredient(s)	Product	Processing technology employed	References
Beverage	Cocoa, coffee, skim milk powder, sugar, vegetable fat/oil, chocolate	Chocolate coated beverage mix	Spray drying	Camp and Fischbach (1990)
	Cocoa powder, liquid binder	Quick dissolving granular cocoa	Granulation in a fluidized bed dryer	Kimura and Teraunchi (1999)
	Cocoa, sugar, maltodextrin, milk	Coated agglomerates	Mixing–agglomeration–coating	Kowalska and Lenart (2005)
	Juice concentrate from vegetable/fruit, whey/soy/pea/rice/nut/egg white/wheat protein	Powdered protein beverage mix	Mixing	Sherwood, Jenkins and Rittmanic (2008)
	Cocoa powder	Readily dispersible cocoa granules	Agglomeration by wet granulation–drying	Vu *et al.* (2003)
	Sugar, cocoa, coffee extract	Dried agglomerates	Jet agglomeration–drying	Schuchmann (1995)
Milk	Milk	Whole/skim milk instant powder	Agglomeration by spray drying	Rennie *et al.* (1999)
	Skim milk, cream, whey powder, sodium caseinate, fat/oil, sugar, locust bean gum	Ice cream mix	Mixing–spray drying	Vega, Goff and Roos (2005)
Miscellaneous	Gum acacia, starch, maltodextrin, oil, esters	Encapsulated flavours	Spray drying–agglomeration in fluidized bed	Buffo *et al.* (2002)
	Concentrated bayberry juice, maltodextrin	Instant bayberry powder	Spray drying–fluidized bed agglomeration	Gong *et al.* (2007)

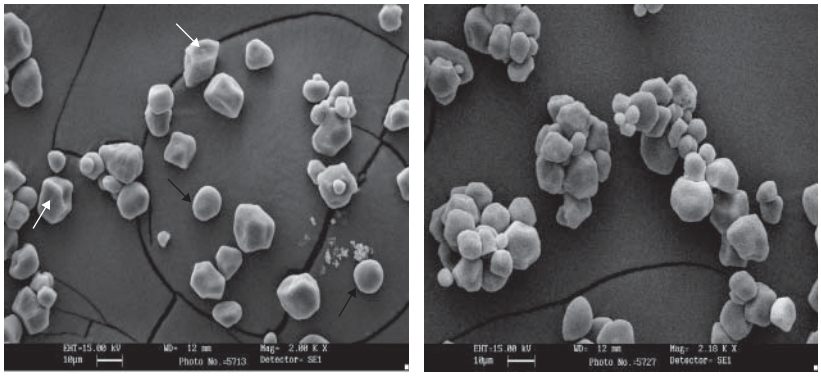

Figure 13.6 Microstructures of raw corn starch (left) and agglomerated (right) corn starch using pre-gelatinized starch. White arrows indicate the flat surfaces of the cuboids while black arrows are for spheroid shapes of raw corn starch, respectively (source: Dhanalakshmi and Bhattacharya 2012. Reproduced with permission of John Wiley & Sons, Ltd.)

Figure 13.7 Unprocessed (raw) and agglomerated corn starch samples. The latter sample is obtained by moistening, steaming and granulation followed by final drying

practice, namely dry and wet granulation. Dry granulation is carried out without the addition of any binder and uses the cohesive nature of the particles and pressure by extruding, tumbling and fluidizing to form larger granules. In a wet granulation process, a binder liquid such as water is most commonly used for the formation of granules (Figure 13.7). The granulation operation may be accompanied by fluidization. Flours of cereals, pulses and beverage powders are some of the agglomerated granular products (Nishii and Horio, 2007).

13.3.2 Drying

Drying is a common operation to obtain instantized products, which mainly include spray drying (Chen and Mazumdar, 2009), freeze drying (Barbosa-Canovas *et al.*, 2005), fluidized bed drying (Tang, Feng and Shen, 2003) and drum drying (Jinapong, Suphantharika and Jamnong, 2008). The process to be carried out is mainly selected based upon the raw material characteristics such as sensitivity to heat, nature of instant product to be obtained and the cost of processing. Baby food powders, ice cream mixes, vegetable and fruit juice powders, coffee powder, whey protein concentrates, soup mixes, egg white and yolk powders are some of the products where drying has an important role.

13.4 Scientific principles

It is desirable that the development of instant products is linked with an understanding of the scientific principles of food preservation and processing including the physical and physicochemical changes in food. As the main target of the instantization process is to reduce the cooking/processing time at the consumer end, providing convenience, obviously a part of the cooking process is done during instantization. The following changes are expected during the instantisation process.

13.4.1 Gelatinization and dextrinization

Starch is the most common carbohydrate polymer present in cereals, pulses, vegetables and tuber crops. As the unprocessed starch is difficult to digest in human systems, the gelatinization process is induced wherein the crystalline structure of starch changes to an amorphous state due to swelling of granules as a result of water absorption. This process is dependent on the moisture content of the product, time and temperature of processing (Kohyama *et al.*, 2004). Continuation of the moist heating process causes bursting of the already swollen granules and the process of dextrinization sets in. The high molecular weight branched structure of amylopectin becomes ruptured during the cooking process. This can be achieved during the process of flaking between two rotary rollers or during extrusion cooking (Ng *et al.*, 1999). Though retrogradation is not a desirable feature in the instantiszation process, it cannot be completely avoided. The popular traditional product like flaked rice is developed by flaking the cooked rice wherein the native structure of starch is severely affected, and thus the chance of retrogradation is low. In turn, the water absorption capacity of dried flaked rice markedly increases such that it can be consumed just after rehydration at room temperature.

13.4.2 Softening

Many cereals and pulses are extremely hard in texture. Reduction in size followed by hydrothermal treatment softens the finished product. Further, vegetables, meat and fish are cooked/roasted to induce softening in addition to developing a desirable flavour/taste to improve palatability.

13.4.3 Changes in protein

The thermal treatments offer a wide range of changes in the structural and functional properties of proteins, including digestibility. For example, thermal treatment improves the digestibility of egg protein (Vijayakumari, Pugalenthi and Vadivel, 2007) in addition to denaturation of proteins and alteration in the water absorption capacity.

13.4.4 Inactivation of antinutritional factors

Thermal treatment can inactivate several antinutritional factors such as protease and alpha-amylase inhibitors present in cereals and trypsin inhibitor, phytic acid, polyphenols, lectin, haemagglutinating activity in pulses/oilseeds such as soybean (Alonso, Orue and Marzo, 1998).

13.4.5 Improvement in palatability and functional characteristics

Improvement in the palatability attributes such as taste and flavour is also accomplished in instant foods like cooked fish/meat (Lewu, Adebola and Afolayan, 2010). The raw or uncooked taste of cereal/pulse/meat/fish is avoided along with suppression of the beany flavour in pulses. The functional attributes, like wetting/hydration, dissolution and dispersability, are improved to offer convenience foods that require less time in preparation.

13.5 Conclusions and future possibilities

Instant food, though not defined in an unambiguous scientific manner at present, is a reality of the present day lifestyle. These foods have been developed out of the need for people possessing less time for cooking in an elaborate manner. The specific advantages of instant foods are reducing drudgery and time of preparation, and inducing improved functional properties like quick rehydration, high dispersability/wettability, improved handling characteristics and possibly nutritional superiority.

Instantization is a composite technology rather than an individual processing technique. It is often coupled with agglomeration/granulation, fast drying/freeze drying and mixing with appropriate ingredients and additives, which are to be carefully selected and applied judiciously in a sequential manner. Application of the agglomeration method needs a special mention in relation to the development of instant foods.

Possibly all sources of raw food ingredients are used for instantization. However, cereals are a more frequently used commodity compared to other ingredients. Currently, most of the instantization processes are covered under patents while the number of research articles is limited. It is hoped that researchers will provide adequate attention and emphasis on such processing systems and products, considering the challenges of making more products convenient to end users as well as for industries focusing on innovations. In addition to products already manufactured, scope exists to develop many more products in the near future. The new product list may include instant jam/jelly, imitation egg and meat products using nonmeat/fish/poultry sources, and possibly instant bread, biscuit and cookies. However, a search for newer ingredients and processing technologies is needed to make these products feasible.

References

Aleksandrovich, R.M. (2001) Instant food. World Patent WO 0189321A1.

Aleksandrovich, R.M. (2002) Instant food product. World Patent WO 0200037A2.

Alonso, R., Orue, E. and Marzo, F. (1998) Effects of extrusion and conventional processing methods on protein and antinutritional factor contents in pea seeds. *Food Chemistry*, **63**, 505–512.

Antwi-Afriyie, M. (2005) Instant food. German Patent DE 10355945A1.

Baker, C.G.J. (1997) *Industrial Drying of Foods*. Blackie Academic and Professional, London, England, pp. 122–124.

Baldwin, A.J. and Sanderson, W.B. (1972) Instant whole milk powder. *New Zealand Journal of Dairy Science and Technology*, **7**, 21–22.

Barbosa-Canovas, G.V., Ortega-Rivas, E., Juliano, P. and Yan, H. (2005) *Food Powders – Physical Properties, Processing and Functionality*. Kluwer Academic/Plenum Publishers, London, England, pp. 175–178.

Berizzi, H.P. (2004) Process for preparing flour mixes for baking purposes. German Federal Republic Patent DE 10248160A1.

Bhuibhar, B.W., Sawant, B.P., Andhare, V.K. and Kadu, A.B. (1991) A study on instantisation of red gram dhal. *Journal of Food Science and Techonology*, **28**, 84–85.

Bohrmann, H., Bezner, K. and Sidler, R. (1979) Process for producing a dry product for food preparations. US Patent 4156020.

Bruemmer, T. (2003) Instant modified flours. German Patent DE 10214644A1.

Buffo, R.A., Probst, K., Zehentbauer, G., Luo, Z. and Reineccius, G.A. (2002) Effects of agglomeration on the properties of spray-dried encapsulated flavours. *Flavour Fragrance*, **17**, 292–299.

Butterbaugh, J.L. and Sargent, J.A. (2002) Dissolution agent and process for making rapidly soluble instant beverage products. US Patent US 6379737B1.

Camp, W.F. and Fischbach, E.R. (1990) Chocolate coated beverage mixes. US Patent US4980181.

Capodieci, R.A. (2002) Cereal food product and method. US Patent US 20020127310 A1.

Chen, X.D. and Mazumdar, A.S. (2009) *Drying Technologies in Food Processing*. John Wiley & Sons, Ltd, Chichester, England, pp. 212–218.

Chen, A.C.C., Lang, C.E., Graham, C.P. and Rizzuto, A.B. (1982) Crystallized, readily water-dispersible sugar product. US Patent US 4338350.

Cremer, B.K., Leung, H.K., Manis, B., Miller, K.S., Niermann, J.T., Root, T.F., Sheppard, M.W., Stalder, J. and Wayne, E. (2008) Free flowing vegetable powder and method for its manufacture. US Patent US 20080182006A1.

Cunningham, C.R. (2007) Agglomerated starch compositions. US Patent US 7186293B2.

Denis, G. and Petra, B. (2006) Instant food for age groups and method for its production. German Patent DE 102005024410A1.

Dewettinck, K., Deroo, L., Messens, W. and Huyghebaert, A. (1999) Agglomeration tendency during top-spray fluidized bed coating with gelatin and starch hydrolysate. *Lebensmittel-Wissenschaftund-Technologie*, **32**, 102–106.

Dhanalakshmi, K. and Bhattacharya, S. (2011) Compaction agglomeration of corn starch in presence of different binders. *Journal of Food Engineering*, **104**, 348–355.

Dhanalakshmi, K. and Bhattacharya, S. (2012) Flow and functional characterization of cornstarch powder in the presence of water and pregelatinized starch. *Journal of Food Process Engineering*, **35**, 887–897.

Eu-Chin, L. (2000) Method and apparatus for making flakes. World Patent WO0044243.

Fairhurst, P., Labbe, D. and Ortega, G. (2002) Soluble black coffee. European Patent EP 1201135A1.

Gallen, V.A. (2002) Instant powder and its use for preparation of instant drinks for use with low-meat or meat-free diets. German Patent DE 10105689A1.

George, G. and Saksena, S. (2002) Cold water soluble tea concentrate. US Patent US 20020197379A1.

Ghosal, S. Indira, T.N. and Bhattacharya, S. (2010) Agglomeration of a model food powder: effect of maltodextrin and gum Arabic dispersions on flow behavior and compacted mass. *Journal of Food Engineering*, **96**, 222–228.

Gong, Z., Zhang, M., Mujumdar, A.S. and Sun, J. (2007) Spray drying and agglomeration of instant bayberry powder. *Drying Technology*, **26**, 116–121.

Groote, E.D., Rainer, P.-E. and Tomas, K. (2006) Instant food product or beverage has powder product applied to sweetener crystals especially sugar and pressed into single dose blocks. German Patent DE 102005017613A1.

Gurkin, M., Bardonia, N.Y., Sanderson, G.W. and Graham, H.N. (1972) Process for making a spray-dried instant tea of desired bulk density. US Patent US 3666484.

Hase, Y., Yamauchi, M. and Handa, R. (1980) Process dried beans as instant food and their manufacturing method. US Patent US 4214007.

Jensen, J.D. (1973) Methods of instantizing powders for the preparation of food drinks. *Manufacturing Confectioner*, **53**, 47–50.

Jensen, J.D. (1975) Some recent advances in agglomerating, instantizing and spray drying. *Food Technology*, **6**, 60–71.

Jinapong, N., Suphantharika, M. and Jamnong, P. (2008) Production of instant soymilk powders by ultrafiltration, spray drying and fluidized bed agglomeration. *Journal of Food Engineering*, **84**, 194–205.

Katt, J.L., Moore, C.O. and Eastman, J.E. (1986) Dry instant food composition. US Patent US 4623549.

Kenke, D. and Walkowiak, R. (2000) Pressed ground coffee or tea leaf tablets disintegrating in aqueous liquid and storable coffee or tea extract tablets contain disintegration agent, wetting agent, additive and gelatin as binder. German Patent DE 19927963A1.

Kessler, U., Duffey, J.L., Dreyer, M.G., Kauter, M.D., Sudarshan, M.B., Chanvrier, H.M.J., Wehrspann, O. and Fu, X. (2010) Instant drink powder. US Patent US 20100215818 A1.

Kimura, Y. and Teraunchi, M. (1999) Process for producing granular cocoa. US Patent US 6007857.

Kohyama, K., Matsuki, J., Yasui, T. and Sasaki, T. (2004) A differential thermal analysis of the gelatinization and retrogradation of wheat starches with different amylopectin chain lengths. *Carbohydrate Polymers*, **58**, 71–77.

Kowalska, J. and Lenart, A. (2005) The influence of ingredients distribution on properties of agglomerated cocoa products. *Journal of Food Engineering*, **68**, 155–161.

Kumlehn, D., Vatterott, W., Batalia, M., Stubbe, H. and Haug, M.E. (2004) Spray-dried soluble instant powder and instant powder beverage for producing beverages and corresponding production method. World Patent WO 2004017761A1.

Kuti, S.A. (1978) Agglomeration – the practical alternative. *Journal of American Oil Chemists' Society*, **55**, 141–143.

Lee, E. and Wissgott, U. (2001) Instant soakable rice. US Patent US 20010006696A1.

Lewu, M.N., Adebola, P.O. and Afolayan, A.J. (2010) Effect of cooking on the mineral contents and anti-nutritional factors in seven accessions of *Colocasia esculenta* (L.) Schott growing in South Africa. *Journal of Food Composition and Analysis*, **23**, 389–393.

Lian, H.P.R. and Toh, T.S. (1998) Preparation of instant glass noodles. US Patent US 5773069.

Liying, G. (2009) Ginseng nutritious instant tea and preparation method thereof. US Patent US 20090162523A1.

Mahajan, P.V. and Chattopadhyay, P.K. (2000) Development of a chemically leavened cereal–legume based instant mix. *Journal of Food Science and Technology*, **37**, 459–464.

Maini, E. and Maini, M. (2011) Process for producing instantaneous cold soluble gelatin and product thereof. World Patent WO 2011018809A1.

Mueller, E. (1986) Instant food. German Patent DE 3503505A1.

Mujoo, R. and Ali, S. Z. (2000) Changes in physico-chemical and rheological properties of rice during flaking. *International Journal of Food Properties*, **3**, 117–135.

Mukherjee, S. and Bhattacharya, S. (2006) Characterization of agglomeration process as a function of moisture content using a model food powder. *Journal of Texture Studies*, **37**, 35–48.

Murata, S. Shimizu, N. and Kokeguchi, S. (1982) Process dried beans as instant foods and their manufacturing method. US Patent US 4333955.

Ng, A., Lecain, S., Parker, M.L. Smith, A.C. and Waldron, K.W. (1999) Modification of cell-wall polymers of onion waste. III. Effect of extrusion–cooking on cell-wall material of outer fleshy tissues. *Carbohydrate Polymers*, **39**, 341–349.

Nishii, K. and Horio, M. (2007) Dry granulation. In: Salman, A.D., Hounslow, M.J. and Seville, J.P.K. (eds), *Handbook of Powder Technology*, vol. 11. Elsevier, Oxford, England, pp. 289–322.

Nove, K.M.-H. (2007) Methods for the preparation of a dry instant food comprising at least two components as well as dry instant food prepared by the method. US Patent US 20100227028.

Palzer, S. (2007) Agglomeration of dehydrated consumer foods. In: Salman, A.D., Hounslow, M.J. and Seville, J.P.K. (eds), *Handbook of Powder Technology*, vol. 11. Elsevier, Oxford, England, pp. 592–646.

Peitsch, W. (1991) *Size Enlargement by Agglomeration*. John Wiley & Sons, Ltd, Chichester, England, pp. 123–413.

Pietsch, W. (2002) *Agglomeration Processes: Phenomenon, Technologies, Equipment*, Wiley-VCH Verlag GmbH, Weinheim. Germany, pp. 133–408.

Pisecky, J. and Westergaard, V. (1972) Manufacture of instant whole milk soluble in cold water. *Dairy Industry*, **37**(3), 144–147.

Quandt, L., Coalson, J.A., Shields, D. and Field, R. (2010) Instantizer and methods of using the same. US Patent US 20100209549A1.

Rennie, P.R., Chen, X.D., Hargreaves, C. and Mackereth, A.R. (1999) A study of the cohesion of dairy powders. *Journal of Food Engineering*, **39**, 277–284.

Ryk, M.A. (2001) Instant food. World Patent WO 0189321A1.

Schechter, S.M. (2000) Textured dry mix instant nutritional drink. US Patent US 6071547.

Schuchmann, H. (1995) Production of instant foods by jet agglomeration. *Food Control*, **6**, 95–100.

Sherwood, S., Jenkins, D.A. and Rittmanic, S.A. (2008) Powdered protein beverage mix and methods of making the same. US Patent US 2008050498 A1.

Steiger, D. (2006) Production of readily wettable, instantized powder for use in foods or feeds, by spraying particles having free surface fat, especially based on coconut milk, with lecithin and alginate in water then drying. German Patent DE 102004038910A1.

Sterzel, H.J. (2001) Instant powder for manufacture of carbonated alcoholic beverages. German Patent DE19948047A1.

Stoltze, A. and Masters, K. (1979) Recent developments in the manufacture of instant coffee and coffee substitutes. *Food Chemistry*, **4**, 31–39.

Strommen, I.. Eikevik, T.M. and Alves-Filho, O. (2001) Method for producing instant powder. World Patent WO 0139616A1.

Takeiti, C.Y., Kieckbusch, T.G. and Collares-Queiroz, F.P. (2008) Optimization of the jet steam instantizing process of commercial maltodextrin powders. *Journal of Food Engineering*, **86**, 444–452.

Tang, J., Feng, H. and Shen, G.-Q. (2003) Drum drying. In: Heldman, D.R. (ed.), *Encyclopaedia of Agricultural and Food Engineering*. Marcel Dekker, Inc., New York, USA, pp. 211–213.

Tuason, D.C. and McGinley, E.J. (1990) Microcrystalline cellulose-based stabilizer system for dry mix instant chocolate drink. US Patent US 4980193.

Turchiuli, C., Eloualia, Z., Mansouri, N.E. and Dumoulin, E. (2005) Fluidised bed agglomeration: agglomerates shape and end-use properties. *Powder Technology*, **157**, 168–175.

Upadhyay, K.G., Vyas, S.H. and Pandya, A.J. (1982) Instantization of milk powders. *Indian Dairyman*, **34**, 187–96.

Vega, C., Goff, H.D. and Roos, Y.H. (2005) Spray drying of high-sucrose dairy emulsions: feasibility and physicochemical properties. *Journal of Food Science*, **70**, E244–E251.

Vijayakumari, K., Pugalenthi, M. and Vadivel, V. (2007) Effect of soaking and hydrothermal processing methods on the levels of antinutrients and *in vitro* protein digestibility of *Bauhinia purpurea* L. seeds. *Food Chemistry*, **103**, 968–975.

Villagran, F.V., Butterbaugh, J.L., Small, L.E. and Sargent, J.A. (2002) Fortified beverages with improved texture and flavour impact at lower dosage of solids. US Patent US 20020037353A1.

Vu, T.O., Galet, L., Fages, J. and Oulahna, D. (2003) Improving the dispersion kinetics of a cocoa powder by size enlargement. *Powder Technology*, **130**, 400–406.

Waskow, M., Ruempler, K. and Jacob, M. (2005) A process for production of instant products in a spouted bed. German Patent DE 102004024681A1.

Yoshitoshi, S. (2006) Artificial shark fin, method for producing the same, and instant food comprising the same. Japanese Patent JP 2006238856.

Youssef, M.M. (1990) Instantization and evaluation of some traditional Egyptian foods. *Food Chemistry*, **38**, 247–254.

Yusof, Y.A., Smith, A.C. and Briscoe, B.J. (2005) Roll compaction of maize powder. *Chemical Engineering Science*, **60**, 3919–3931.

Zameitat, W. (1976) Production of instant food in flake form. German Patent DE 2457647 A1.

Zeller, B.L. (1998) Foaming coffee creamer and instant hot cappuccino. US Patent US 5721003.

Zhao, J.J. and Bertrand, D.T. (2004) Instantly dispersible pregelatinized starches for use in food products. US Patent US 2004258831A1.

14

Fortification and Impregnation Practices in Food Processing

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14.1 Introduction

The supply of processed products in the food sector increases in combination with consumer demands on the single products. With a view to the food fortification aspects, there are three main perspectives that will be of interest. The first is the global demand for functional foods with a wide range of fortification possibilities. The second one is the consumer demand for minimally processed food, which generates new approaches in food modelling. Lastly, there are new perspectives in the food technology field, due to the combination of well-known industrial technological processes. The aim of the combination strategy is to develop new products or processes using the advantages of approved production methods.

In general, food composition is not only influenced by the selection of raw materials and their production methods such as cultivation conditions, breeding and storage but also by food fortification techniques like atmospheric and vacuum impregnation (VI) or osmotic dehydration (OD). These applications are established methods and are used for different foods, especially for porous raw materials. The main applications for OD are salting and brining of cheese, fish and meat or the controlled modification of vegetable and fruit compositions by water removal. The aim of this chapter is to provide an overview

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on OD and VI in food with the focus on process influencing parameters, and recent and the near future applications.

Fortification generally aims to prevent or correct a deficiency of nutrients like vitamins, minerals or other health promoting substances. The technological challenges of food fortification are the modification of the physico-chemical properties of a particular active ingredient, the interactions between an additive and the food matrix, the uptake, translocation properties, the bioavailability, the stability as well as sensory aspects like taste and colour changes. Fortifications for human health are carried out with micro- and macronutrients and other value-giving ingredients such as fibres or secondary plant products. Many foods and beverages worldwide have been fortified with micronutrients like minerals (zinc and iron), vitamins (vitamin A, vitamin D and folic acid) or iodine. Examples of food macronutrient fortifications are mixing of solid powders such as addition of high protein sources in cereal powders or the encapsulation of omega-3 fatty acid rich fish oil. The wide range of fortification processes depends on the product type and the physicochemical properties of the additive. For dry foods like cereal flour, milk and beverage powders, the pre-blended nutrient mixtures are used. Further fortification practices are the encapsulation and coating technologies for sensitive additives, and the homogenization of liquid food products.

14.2 Food modification by vacuum impregnation

Vacuum impregnation (VI) is a food fortification process that is characterized by the mass transfer between an external impregnation solution and the food matrix as a result of the exposure to vacuum. In contrast, atmospheric impregnation is a fortification process without pressure changes, which is characterized by simple impregnation and mass transfer initiated by immersion and equilibrium reactions.

During VI, the porous fraction of food is penetrated by external liquids under controlled conditions. The process can be divided in the vacuum phase, when subatmospheric pressure is applied, and the atmospheric phase, when the atmospheric pressure conditions are restored and the food is still immersed in the VI solution. Figure 14.1a represents a porous food; this tissue model consists of cells and the intercellular spaces. Figure 14.1b demonstrates the immersion of the porous cell complex in a liquid under atmospheric pressure. Close to the surface of the model tissue, a liquid influx can be observed. The only driving force for this mass transfer is the capillary pressure. The interaction between the raw material and the impregnation solution is limited to the outer parts of the food matrix. Under exposure to vacuum conditions (Figure 14.1c), the gas of the porous tissue expands and flows out in combination with the deformation relaxation phenomena (DRP) of the solid. This vacuum phase is characterized by the duration and pressure

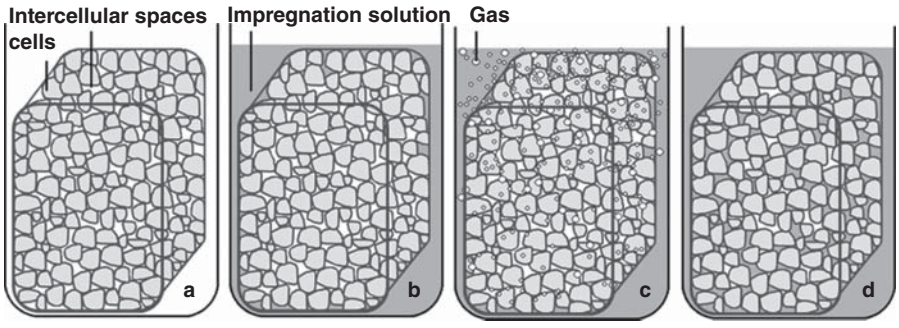

Figure 14.1 Schematic representation of the liquid and gas exchange steps during the vacuum impregnation process of a porous cell complex (a) Native plant tissue; (b) Plant tissue soaked in impregnation solution under atmospheric pressure conditions; (c) Gas release from intercellular spaces during the vacuum phase; (d) Vacuum impregnated plant tissue soaked in impregnation solution after restoring the atmospheric pressure conditions.

level. Investigations of different vegetables showed that a duration of 5 to 15 min was enough to degasify tissues of different porosities (Gras *et al.*, 2002). The plant material responds to the pressure gradient with viscoelastic behaviour (Zhao and Xie, 2004) so that plant tissue swelling occurred during the vacuum phase of the VI process (Chiralt and Fito, 2003). Restoration of the atmospheric pressure (Figure 14.1d) caused air compression and an incorporation of the VI solution in the previously air-filled spaces (Guillemin *et al.*, 2006). The tissue contracts and enhances the soakage of the VI solution (Guillemin *et al.*, 2008; Fito *et al.*, 1996). The positive pressure gradient acts as the driving force. These mass transfer phenomena are described as a hydrodynamic mechanism (HDM). The volume of externally penetrating liquid could account for almost the total volume of the whole previously air-filled spaces (Fito, 1994).

14.3 Food modification by osmotic dehydration

Since the 1960s, osmotic dehydration (OD) has traditionally been used as an industrial preservation method. Beyond the primary goal, which is the water loss of cells by osmotic pressure, various modifications of the food matrix in the process chain are required today on the basis of equilibrium reactions between the raw material and a highly concentrated osmotic solution.

OD is a mass transfer mechanism that occurs if cellular material comes into direct contact with a concentrated hypertonic solution. The medium contains highly concentrated sugar (candying) or salt (salting) concentration, which both control and trigger the transfer processes. Sucrose is a common osmotic agent used for plants, especially for fruit applications. Sodium

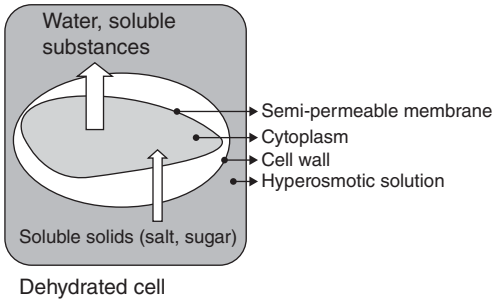

Figure 14.2 Transport and regulation processes in food cells during osmotic dehydration

chloride solutions are primarily used for salting of cheese, meat, fish and vegetables. By the immersion of product tissue in a concentrated solution, different selective mass transfer mechanism between the cells and the OD solution occurs. The two most important transportation processes are the partial water loss and countercurrent osmotic solute uptake of the treated product. In addition to the water removal, dissolved ingredients of the product are released from the cells. These interactions are based on molecular diffusion through the semi-permeable cell membrane. A plasmolysed plant cell with the schematic principles of mass transfer mechanism during OD is shown in Figure 14.2. The arrows indicate the direction of movement caused by equilibrium reactions, which result in the passive outflow of water and soluble ingredients (minerals, vitamins, sugars and acids) from the cell in combination with the influx of the hyperosmotic solution with salt or sugar molecules. The hyperosmotic solution also offers opportunities for the product modification by incorporation of further substances. The cell response and water loss leads to shrinkage of the cytoplasm. Finally, the cell dehydration is a function of the concentration of hypertonic solution. The tolerance to deplasmolysis of the cells is related to the osmotic stress level. An apoptotic cell death may be the result of excessive osmotic cell shrinkage and an osmotic shock. In general, the outcome of OD applications are changes of cell turgidity, cell elasticity, cell viability, tissue firmness, the moisture/solute ratio and the alteration of cell resistance and mechanical behaviour as well.

14.4 Influence parameters on food modification by VI and OD

The principles of food modification possibilities by VI and OD can be divided into three sections: process conditions, product characteristics and solution characteristics (Figure 14.3). Process conditions include the main

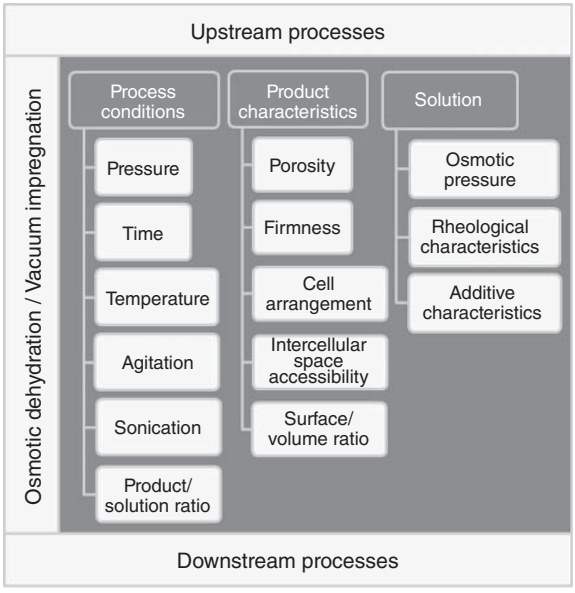

Figure 14.3 Overview of process management factors to control the food composition by osmotic dehydration or vacuum impregnation applications

system parameters, which are responsible for the circumstances of treatment. The product characteristics contain the material properties and quality changes by potential previous upstreaming processes. Third and lastly, the solution composition is listed. With such tools in hand, scientists and food technologists may be able to modify and control the process of OD and VI to modulate food for specific industries and consumer demands.

14.4.1 Process conditions

Pressure The pressure conditions directly influence the mass transfer process during VI and OD. Traditionally, the OD treatment takes place under atmospheric pressure conditions whereas the VI process is based on subatmospheric pressure levels. There are some exceptions to the described pressure settings. Vacuum osmotic dehydration (VOD) is OD under vacuum treatment and the so-called atmospheric impregnation (AD) is a performed impregnation process without vacuum treatment. There are two main reasons for the pressure application during VI; first, the stronger the adjusted vacuum, the stronger is the air outflow of porous tissues, which affects the penetration depth of the impregnation solution after restoring the atmospheric pressure conditions. Usually, vacuum levels between 5 and 200 mbar are applied. Analysis concerning the influence of pressure on apple

slices showed a significant increase in the impregnation rate by lowering the vacuum pressure (Schulze *et al.*, 2012). Therefore, the authors controlled the impregnation process (vacuum between 100 and 800 mbar) by measuring the weight increase and the concentration of impregnated plant extract. Most of these studies worked with a vacuum of 100 mbar. Lower pressure increases the contribution uniformity of the VI solution in the impregnated tissue (Guillemin *et al.*, 2006). These authors reported that the enzyme distribution after vacuum treatment was more homogeneous compared to soaking experiments. Furthermore, the application of vacuum in OD and VI has functional importance for oxygen-dependent reactions. Because oxygen is removed from the product under vacuum conditions, the oxidation reactions can be interrupted. One such example is the undesired enzymatic browning of fruit samples.

Time Another important parameter is the duration of the VI or OD treatment. The duration differs substantially between these two processes and is moreover strongly product specific. The total time in VI can be further divided into the duration of the vacuum phase and the duration of the subsequent atmospheric phase in which the product is inserted in the solution. The duration of the vacuum phase is usually 2.5 to 25 min, but for some VI treatments having longer exposure times up to 60 min can be applied. In the majority of VI procedures, the duration of the atmospheric pressure phase exceeds the vacuum phase. In summary, the listed references in this chapter showed atmospheric pressure phase durations between 5 and 60 min. The time selection is mainly based on the product type and the chosen solution characteristics. The combinations of pressure and time of the two phases open up a scope for quite different impregnation methods and products. The process of VI allows a product treatment in a short time while an OD process takes longer. For example, brining manchego cheese by OD took 18 h whereas the duration of VI treatment accounted for 60 min at a vacuum pressure of 37 mbar as well as 60 min for the atmospheric pressure phase (Guamis *et al.*, 1997).

Temperature A basic influencing factor on product quality and the mass transfer is the process temperature. One argument for the use of OD to reduce the water content when compared to drying methods is that the treatment can be done at lower process temperatures. This is accompanied by the lower loss of heat-sensitive ingredients like vitamins or for the sensory important substances like volatile flavour or colour components. Discrepancies between the used temperature conditions can be attributed to the product sensitivity. Thus, mild conditions at 20 up to 40 °C are applicable mainly for heat-sensitive products; however, higher solution temperature ranges could be used, as long as the desired product characteristics are significantly unaffected. Barat, Chiralt and Fito (2001) characterized the effect of temperature on the mass transfer

mechanism. The kinetic influence of temperature was well predicted by the Arrhenius equation.

Agitation Continuous agitation of samples during the process in comparison to unmoved products can enhance the extent of mass transfer. Mavroudis, Gekas and Sjöholm (1998) observed a correlation between the Reynolds number that is attributed to the agitation level and the water loss during OD. The mass transfer was quite different for the solid gain, which was not significantly affected by the degree of agitation. Moreover, Azuara, Garcia and Beristain (1996) measured the influence of centrifugal forces on OD for apples and potatoes during the OD treatment; the water loss was enhanced by the centrifugal forces. However, in comparison to static OD, the absorption of solids from the osmotic solutions was reduced.

Sonication Besides agitation, the use of ultrasound can have a supporting role in the mass transfer mechanism. Simal *et al.* (1998) reported a supporting role of sonication during OD of fruits. The combination of ultrasonic application and OD increased the transfer rates. Both the water loss and the solute gain rates were enhanced. The authors concluded that ultrasound is more effective for the use of agitation during the OD process. One explanation for the increased mass transfer by ultrasonic wave application was given by Fernandes, Gallão and Rodrigues (2008). They argued that the ultrasound application resulted in the formation of microscopic channels, which increased the water diffusivity in the product.

Product/solution ratio The last type of listed process parameters that influences the OD and VI is the product/solution ratio. The product/solution ratio alters during the process due to the gain of solids into the sample, the antagonistic loss of water and the loss of dissolved substances in the treated product. For this purpose, the product/solution ratio must be set high enough to rule out the possibility of influencing the process. In other words, for OD and VI studies with the aim to calculate mass transfer process models, the solution volume should be selected according to the respective product volume. Bilbao-Sáinz, Andrés and Fito (2005) and Barat, Chiralt and Fito (2001) chose a product/solution ratio of 50:1 for their OD and VI experiments, respectively. For simple applications, the minimum ratio should be selected in such a way that the products are at least covered completely with used OD and VI solutions.

14.4.2 Product characteristics

According to the process studies, one important part of research on OD and VI has focused on the product characteristics. The results from these investigations showed that liquid penetration and mass transfer processes are affected

by several factors, including the product type, species, ripening stage, texture and tissue characteristics, etc. Differences of the used product were not only influenced by the choice of raw material but also by possible pre-treatments, which are responsible for additionally generated changes of the product characteristics.

Porosity Porosity studies demonstrate that the level of gas inside the raw material is directly involved in the penetration degree of the solution in the VI process. The porosity influences the gas outflow of the porous tissues during the vacuum phase in VI (Andrés *et al.*, 2001). The more gas flows out of the tissue, the higher is the possible replacement by the liquid phase. The effective porosity describes the volume of air that can be exchanged by the solution inside the tissue (Zhao and Xie, 2004). It is an important parameter to explain the behaviour of different tissues during the VI process. The modification of air and liquid volume fractions, for example, influences the mechanical behaviour. Salvatori *et al.* (1998) monitored the VI level of different fruits and the impact of the porosity and the effective porosity to HDM and DRP mechanisms. They showed that the structural swelling during VI depends on the initial porosity and therefore has an effect on the HDM, the effective porosity and liquid penetration. These suggest that characterizing the porosity is helpful for the choice of the raw material. Porosity strongly differs between raw material type (cheese, meat, fish, vegetables and fruits), variety, quality and maturity level of tissue. The parenchyma of the apple cortex is a plant tissue with high porosity and is ideal for VI (Guillemin *et al.*, 2006). Thus, apples are often used as model food to study the VI process. From the centre to the periphery of the apple parenchyma, the porosity and intercellular spaces increase and the pore volume elevates during apple growth (Mendoza *et al.*, 2010). VI treatment of apple slices demonstrated an influence of the porosity levels. Schulze *et al.* (2012) reported that for the inner apple parenchyma, a higher VI liquid uptake occurs compared to the porous outer tissue. Carrots, as nonporous vegetables, could also be impregnated successfully (Gras *et al.*, 2002). Indeed, porosity is an important raw material attribute. However, further studies on histological and cellular characteristics are needed.

Firmness The sample texture and the resulting firmness affect the characteristics of deformation because of relevant pressure changes during VI. The relationship between the firmness of 14 different apple cultivars and the VI success was analysed by determining the polyphenol content and weight changes after VI application (Schulze *et al.*, 2012). In conclusion, the investigations showed a significant relationship between the fortification level and fruit firmness. The softer the apple variety, the higher is the resulting polyphenol content (Schulze *et al.*, 2012).

Research data of Torreggiani and Bertolo (2001) about the influence of the ripening stage and firmness on the OD showed an impact on the mass transfer process of kiwi fruit slices. The authors measured a stronger soluble solid gain and a decreased water loss for softer kiwi slices compared to the firmer fruits. After the additional freezing and thawing, as a second and third process step, the texture differed markedly (Torreggiani and Bertolo, 2001).

Cell arrangement The cell arrangement in fruits and vegetables is influenced from a histological point of view and the heterogeneity of cell tissues between the cultivars. Even if the same raw material could be taken for OD and VI investigations, the behaviour during the process may vary. Studies of Mendoza *et al.* (2010) on cell morphology of apple tissues showed cell anisotropy in both the variety and different cultivars. In addition to cell anisotropy, the cell and pore arrangements depend on the apple variety and its growth (Mendoza *et al.*, 2010). Furthermore, the cell morphology and cell from the same apple showed structural differences; in fact, the inner part differs from the outer regions of the apple cortex (Verboven *et al.*, 2008; Khan and Vincent, 1990). With respect to the tissue structure and cell arrangement, it could be shown that these large numbers of raw material variants resulted in different impregnation levels (Schulze *et al.*, 2012). The authors demonstrated that the VI levels of different apple varieties varied. Moreover, it was possible to characterize the differences of the solution distribution in the same sample, with respect to the inner and outer apple cell arrangement, by X-ray analysis of the pore structure.

Intercellular space accessibility The food modifications by VI and OD are not only affected by the level of porosity but also by the size and shape of the pores and their distribution. In modelling experiments about VI influencing parameters, Fito *et al.* (1996) suggested that in addition to the size and shape of the gas-filled pores, the communication between the intercellular spaces is to be taken into account for the VI results. Gras *et al.* (2002) described the penetration distribution of several VI solutions into vegetable tissues of beetroot, carrot, eggplant, zucchini, mushroom and oyster mushroom, and their structural characteristics. The structural findings with the arrangement of cells and gas-filled structures resulted in differences in the deformation reactions during pressure changes and were associated with different incorporation levels of the VI solutions. In some tissue types, the total gas volume of the vascular system was not efficiently penetrated with the used liquids (Gras *et al.*, 2002).

Surface/volume ratio Sample size and shape affect the surface/volume ratio and the OD and VI results because the replacement mechanism starts at the surface of the product. In VI, the gas outflow during the vacuum phase depends strongly on the tissue shape of the impregnated food sample. Both

geometry and thickness influence the penetration depth. This means that the thicker a product sample, the more unfavourable the surface/volume ratio and the more difficult is the penetration into deeper tissue parts by the OD or VI solutions.

14.4.3 Solution characteristics

One of the most interesting influencing parameters on the outcome of the mass transfer mechanism is represented by the selected composition of the OD or VI solutions. Previous investigations examined particularly the osmotic pressure (i.e. hyper-, iso- and hypotonic solutions) and the rheological characteristics of the treatment fluid. The basic demands for both the OD and VI solutions are that they are classified as nontoxic and with sensory acceptability. Even the recycling ability by re-using the solutions for several applications is an interesting point when looking for the most appropriate OD or VI solution. In OD treatments, a dewatering of the hyperosmotic solution occurs as well as leaching of additional tissue ingredients. These characteristic changes limit the number of re-use cycles in OD. For VI, a similar recycling behaviour is required, because the mass transfer induced a shift in the solution composition, which consequently may have an impact on the success of VI. The approach for an effective solution management is to achieve the right balance between several issues. First, the economic one includes time and cost management of direct personnel and raw material costs. Second, there is the guarantee of the product quality with respect to vitamin and mineral contents, sensory quality like colour changes or technological needs like the drying behaviour for post-processing steps. A third point is to ensure the effective running of the process, despite the influences from recycling steps. As a last remark, the guarantee of product safety with regard to microbial aspects should be mentioned.

Osmotic pressure The mass transfer processes during OD and VI are substantially influenced by the constitution of the VI solution and the osmotic pressure of the fluid. Basically, the osmotic pressure of the solution can be divided into three groups: hypertonic, isotonic and hypotonic fluids. Some examples of common OD and VI solutions are shown in Table 14.1. In VI, the listed three solution types can be used; in OD, highly concentrated sugar, sugar alcohol or salt containing fluids were applied. Often the solutions contained the disaccharides (sucrose and maltose) or the sugar alcohol (sorbitol), which was diluted in water (Fernandes, Gallão and Rodrigues, 2008; Cárcel *et al.*, 2007; Torreggiani and Bertolo, 2001). Sodium chloride solutions are common for OD salting applications (Azuara, Garcia and Beristain, 1996). There exists a wide concentration range of the osmotically active ingredients in the study designs, even if the raw material characteristics are approximately equal. Azuara, Garcia and Beristain (1996) examined the influence of ultrasound and

Table 14.1 Overview for OD and VI solution types

Solution type	Raw material	Reference
Hypertonic solutions		
Sucrose solution	Fruit	Barat *et al.* (2002); Fernandes, Gallão and Rodrigues (2008)
Sodium chloride solution	Vegetable	Azuara, Garcia and Beristain (1996)
Sodium chloride solution	Cheese	Guamis *et al.* (1997)
Fruit must solution	Fruit	Martínez-Monzó *et al.* (1998)
High fructose corn solution	Fruit	Xie and Zhao (2003)
Isotonic solutions		
Fruit juice	Fruit	Andrés, Bilbao and Fito (2004); Schulze *et al.* (2012)
Sucrose solution	Fruit	Gras *et al.* (2002); Fito *et al.* (2001)
Sucrose solution	Vegetable	Gras *et al.* (2003)
High fructose corn solution	Fruit	Xie and Zhao (2003); Park, Kodihalli and Zhao (2005)
Sodium chloride solution	Mushroom	Fito *et al.* (1996)
Iron gluconate solution	Vegetable	Fito *et al.* (2001)
Calcium gluconate solution	Vegetable	Fito *et al.* (2001)
Hypotonic solutions		
Pectin solution	Fruit	Martínez-Monzó *et al.* (1998)

OD with 45% and 70% sucrose solutions, and 15% and 30% sodium chloride solutions.

Instead of the induced water loss by OD, VI solutions are mainly composed in such a way as to result in an intake of soluble compounds into the raw material. Apple juice is a common external solution for an isotonic VI liquid, especially for apple parenchyma enrichment (Martin-Esparza *et al.*, 2006; Contreras *et al.*, 2005; Andrés, Bilbao and Fito, 2004). One reason for the frequent uses of isotonic VI solutions is the low shrinkage and deformation changes of the VI products, which were described by Zhao and Xie (2004). Besides this advantage, it is often necessary to use isotonic solutions. In order to investigate the influence of further process factors, VI has to be adjusted for the impact of osmotic pressure. In addition to fruit juices other liquids containing sodium chloride, sucrose, honey, high fructose corn syrup or trehalose are usually used, which can be enriched for a specific purpose with further ingredients like acids, vitamins, minerals and enzymes (Derossi, de Pilli and Severini, 2010; Phoon *et al.*, 2008; Guillemin *et al.*, 2006; Lin *et al.*, 2006; Xie and Zhao, 2003; Fito *et al.*, 1996). Besides the application of isotonic solutions, hypo- or hypertonic liquids have also been used in different studies. Hypotonic VI solutions contain only minor amounts of dissolved substances, if any. The water diffuses into the cells, which leads to the swelling and possibly lysis of

the vacuum impregnated tissues. Thus, a shorter processing period of the trials may be beneficial.

VI with hypertonic solutions resulted in additional OD reactions of the cells, like plasmolysis. In apples, VI with 35°, 50° and 65°Brix grape must solutions led to product characteristics like a decreased cell turgidity as well as the loss of the cell elasticity (Martínez-Monzó *et al.*, 1998).

Rheological and additive characteristics In addition to osmotic pressure, rheological properties of the VI solutions play a crucial role for the impregnation level. For VI in plant tissues, often pectin dispersions, for example as cryostabilizers, were used. VI solutions with a high viscosity show a lower transfer rate and can be less incorporated in the porous tissue compared to low-viscosity solutions (Zhao and Xie, 2004). To compensate for these counteracting effects of higher viscosity solutions, it is necessary to extend the time under atmospheric pressure after the vacuum phase (Martínez-Monzó *et al.*, 1998). Another possibility is a hypotonic basis as the VI solution for enhancement of the transfer rate and an improved VI result (Guillemin *et al.*, 2008).

Besides the additive influences on the rheological properties, such as the mentioned pectin, the success of the incorporation depends on further additive characteristics. Especially for food fortifications by VI, structural properties of the additives may result in a different accumulation behaviour. This can be seen, for example, in a study by Lin *et al.* (2006) with the aim of nutritional enrichment of three tocopherol sources in pears. The measured vitamin E concentration after VI depended on the tocopherol source. As an explanation, these authors hypothesized that an interaction in the intercellular spaces might have influenced the incorporation success by restricted movement.

14.5 Traditional and future applications

Possible fields of applications for OD and VI include traditional aspects like stability, preservation and shelf-life of food as well as technological influences on industrial operations. In this context, attributes like firming and texture characteristics and the field of upgrading product characteristics such as health or sensory aspects are of special interest. International organizations such as the World Health Organization (WHO) and Food and Agriculture Organization of the United Nations (FAO) promote a diet rich in vegetable and fruit because of the well-known protective effects on human health. OD and VI could play a useful role to reach these recommendations. In the fruit and vegetable market, OD and VI treatments can provide new opportunities in respect to shelf-life properties and sensory characteristics. Furthermore, they offer new product development strategies in the growing food sectors of functional and ready-to-eat foods.

Food preservation can be realized in many ways, for instance with thermal processes such as pasteurization, sterilization, drying and freezing or other procedures like smoking, sulfiding, pH reduction, brining, candying and the use of preservatives, pressure, irradiation or storage under special conditions (humidity, temperature and atmosphere). The application of OD or VI enables a support in terms of water activity and water content reduction. A particularly crucial point is thereby the possibility of preservation without negative thermal influences, especially for heat-sensitive food ingredients. A high environmental concentration of salt or sugar lowers the water activity level, which is responsible for the preservation effect and can additionally be utilized by vacuum treatment. Therefore the VI technique was used for food preservation like salting cheese (Guamis *et al.*, 1997) and meat (Chiralt *et al.*, 2001) or the candying process of fruits (Barat *et al.*, 2002). The VI during the candying process had supportive effects and resulted in a considerable saving of time within the production process and better sensory properties because VI allowed working with lower process temperatures (Barat *et al.*, 2002). Acidification is an ancient preservation method, usually applied in vegetable tissues. The application of VI improves the acidification process by lowering the pH using lactic acid in red pepper (Derossi, de Pilli and Severini, 2010).

These subatmospheric pressure levels during the acidification process resulted in firmness changes (Derossi, de Pilli and Severini, 2010). In addition to a specific utilization of this effect on firmness, VI may also be used for structural influence by the addition of agents for structural changes. For this purpose, VI with cryostabilizers preserve the fruit quality of frozen storage apples (Martínez-Monzo *et al.*, 1998). Adding cryoprotectant agents to a VI solution resulted in structural changes and cell wall stabilization (Martínez-Monzo *et al.*, 1998). Further texture and firming applications for OD and VI offered the infusion of pectinmethylesterase, high fructose corn syrup and calcium chloride. For technological needs, the firmness of pasteurized fruits could be improved by VI with pectinmethylesterase and calcium chloride (Guillemin *et al.*, 2006). Calcium fortification by VI allows structural changes, because it interacts with the cellular matrix (Gras *et al.*, 2003) as well as the incorporation of high fructose corn syrup, which increased significantly the firmness of apple tissue after the impregnation process (Park, Kodihalli and Zhao, 2005).

Generally, the wide range of OD and VI applications also affects the sensory characteristics of a food product. The above-mentioned salting, candying or acidification processes obviously have an impact on the taste of the treated product. However, apart from that, sensory aspects like browning inhibition in cut fruits and vegetables could be valuable reasons for OD and VI treatments. The enzymatic browning reaction can be considerably decreased by the degasifying step during the vacuum phase and by adding antibrowning agents in the treatment solution. As an effective antibrowning agent in pears and apples, honey could be used in VI (Lin *et al.*, 2006; Jeon and Zhao, 2005). The

inhibition of browning reactions is an opportunity for VI application for new products in the health-oriented ready-to-eat and fresh-cut fruit market sector.

Functional food contains ingredients that offer an additional health promoting benefit besides its nutritive value. In the worldwide growing functional food sector, foods are enriched with probiotics, vitamins, minerals and secondary plant products. In recent years, VI has become a popular method for enrichment. For example, the fat-soluble vitamin tocopherol was impregnated into apples and pears (Lin *et al.*, 2006; Park, Kodihalli and Zhao, 2005). In addition, vitamin and mineral fortification or a combination of both could be performed (Park, Kodihalli and Zhao, 2005; Xie and Zhao, 2003). Furthermore, VI solutions with secondary plant products or probiotics were impregnated in the raw material (Schulze *et al.*, 2012; Betoret *et al.*, 2003). Schulze *et al.* (2012) used polyphenols of apple peel as secondary plant products, which were obtained from pomace as a by-product of the beverage industry.

The utilization of by-products can generate additional benefits for the OD and VI application techniques, because the addition of newly extracted ingredients is an effective and innovative means to achieve this. Recycling of by-products in the food industry to reduce the environmental impact is becoming increasingly important. Thus, this strategy offers a wide range of applications for an efficient and sustainable food industry in the future.

14.6 Combination of OD and VI with other processes

A general trend in the application of the two minimal processing options OD and VI is the combination of several process operations. This possibility makes OD and VI of interest for many food technology operations and product development.

14.6.1 Pre-treatment processes before OD and VI

To improve the industrial potential of OD and VI, pulsed electric fields (PEF) as a preprocessing step were investigated by a small number of studies. Cell disintegration enhanced transfer rates in the OD of red pepper bells. With a view to VI, Phoon *et al.* (2008) successfully improved the freezing tolerance of spinach leaves by using trehalose as a cryoprotectant along with PEF.

Besides the influence of PEF as pre-processing technique, the use of ultrasound and centrifugal forces were studied. Cárcel *et al.* (2007) showed that the osmotic treatment of apples with hypertonic sucrose solutions in combination with ultrasound resulted in enhanced mass transfer. Because of ultrasonication, microscopic channels for water release were formed, which induced an increased water diffusivity of plant tissue (Fernandes, Gallão and

Rodrigues, 2008) The approach of supporting the mass transfer by centrifugal forces was investigated for apple and potato samples (Azuara, Garcia and Beristain, 1996). The results of this study demonstrated an increased water release level due to the combined approach compared to the sole OD application.

14.6.2 Downstream processes after OD and VI

Drying procedures are common downstream processes after OD or VI, especially for vegetable and fruit products. Several drying techniques can be combined with OD and VI. After VI the drying behaviour or product characteristics were investigated with air drying (Contreras *et al.*, 2005; Betoret *et al.*, 2003) or the coupling of convective air drying with microwaves (Bilbao-Sáinz, Andrés and Fito, 2007; Contreras *et al.*, 2005; Andrés, Bilbao and Fito, 2004). All of these studies used apple as a porous food matrix because it acts as a suitable sample material and are well tested in VI examinations. The studies showed that pre-processing of food with VI influenced the drying behaviour, such as the drying duration, the drying efficiency and food quality aspects. This will make it necessary to review previous drying methods and alter the drying process management. Andrés, Bilbao and Fito (2004) analysed VI influence on the drying result with hot air microwave dehydration. The authors reported a higher shrinkage of VI apple samples compared to non-impregnated material. They concluded that this observation was based on the porosity decrease and cell deformation reactions by VI. In comparison, trials of VI apples treated by Contreras *et al.* (2005) showed that the product characteristics differed between the hot-air dried and the microwaves dried samples. After microwave drying, the products resulted in a harder texture but showed a better rehydration behaviour. The authors observed differences in the glass transition temperature and the soluble pectin fraction, which could be responsible for the product properties. With a view to the dielectric properties that are responsible for heating under microwave conditions, Martín-Esparza *et al.* (2006) could demonstrate influences between the modified properties of VI products and the electromagnetic field. Depending on the VI solution, the mobile water fraction in impregnated products increased because of the liquid penetration. According to the dielectric properties, the pattern of product heating changed depending on the level of incorporation and the concentration of additional ingredients.

Often the application of OD is used as a first step of water reduction without thermal stress for the heat-sensitive products. Later, the residual water can be removed by the downstream drying method. Besides the application of drying combinations for extended shelf-life, coupling OD and VI with Ohmic heating treatment can increase the storability. For example, Moreno *et al.* (2012) showed that a treatment of electric field strength of 13 and 17 V/cm after VI

inhibited the growth of microorganisms like moulds, yeasts and mesophiles in microbial-sensitive strawberry samples.

Freezing is a common preservation method, especially for fruits with a short harvest season. Freezing of plant tissue is accompanied with modified fruit composition and cellular damages because of ice crystals, which resulted in texture loss of the thawed products. The applications of OD as well as VI as pre-processing steps to reduce the mentioned disadvantages of freezing were investigated in several studies. So called dehydrofrozen products are characterized by a dewatering step before freezing. In addition to various drying procedures, research on OD application was carried out. Torreggiani and Bertolo (2001) reported a significant retention of chlorophyll and vitamin C after frozen storage due to OD treatment with maltose solution. This also involves an intensive green colour development compared to kiwi fruit slices without OD pre-treatment. Whereas OD is applied to reduce the water content before freezing, VI could be used to decrease cellular damages by the insertion of cryostabilizers and cyroprotectants into VI solutions (Martínez-Monzó *et al.*, 1998). In this study, apples were vacuum impregnated with pectin and sugars from concentrated grape must, which resulted in reduced crystal damage by freezable water and an improved fruit resistance.

14.7 Conclusions

The role of OD and VI applications on food modification has been intensively studied during the past 20 years. Whereas in the beginning of OD and VI research technological benefits and the development of the processes were considered, scientific research has shifted to the development of sophisticated food products. In general, the possibilities to influence food with OD and VI treatments by incorporation of health promoting ingredients are an interesting approach in the field of functional food development. However, there is also a challenge to new requirements in food shelf-life and storage behaviour. Especially in the field of ready-to-eat snacks, the consumption of fruit and vegetable products is becoming more and more a combination of health benefits in association with growing demands from consumers to sensory aspects. The quality impression of a value-adding component in fresh ready-to-eat products, snacks, cereals or other products becomes more popular in the premium food segment in which OD and VI application represents possibilities for satisfying the increased needs of the customers. For creating food with sophisticated properties, the first step is to define the final product exactly by using of a top-down approach. Based on these considerations, the process, the raw material and the solution characteristics may be chosen and combined with one another to ensure the desired result in a pilot scale. The wide range of influencing parameters provides several approaches for product solutions. Finally, to complete the product development, a scale-up with adjustment for

the defined product quality characteristic with fine tuning in the production process is necessary.

Abbreviations

AD	Atmospheric impregnation
DRP	Deformation relaxation phenomena
FAO	Food and Agriculture Organization
HDM	Hydrodynamic mechanism
OD	Osmotic dehydration
PEF	Pulsed electric fields
VI	Vacuum impregnation
VOD	Vacuum osmotic dehydration
WHO	World Health Organization

References

Andrés, A., Bilbao, C. and Fito, P. (2004) Drying kinetics of apple cylinders under combined hot air-microwave dehydration. *Journal of Food Engineering*, **63**(1), 71–78.

Andrés, A., Salvatori, D., Albors, A., Chiralt, A. and Fito, P. (2001) Vacuum impregnation viability of some fruits and vegetables. In: P. Fito, A. Chiralt, J.M. Barat, W.E.L. Spiess and D. Behsnililan (eds), *Osmotic Dehydration and Vacuum Impregnation*. Applications in Food Industries. Food Preservation Technology Series, Technomic Publishers, Lancaster, Pennsylvania, pp. 53–60.

Azuara, E., Garcia, H.S. and Beristain, C.I. (1996) Effect of the centrifugal force on osmotic dehydration of potatoes and apples. *Food Research International*, **29**(2), 195–199.

Barat, J.M., Chiralt, A. and Fito, P. (2001) Effect of osmotic solution concentration, temperature and vacuum impregnation pretreatment on osmotic dehydration kinetics of apple slices. *Food Science and Technology International*, **7**(5), 451–456.

Barat, J. M., Talens, P., Barrera, C., Chiralt, A. and Fito, P. (2002) Pineapple candying at mild temperature by applying vacuum impregnation. *Journal of Food Science*, **67**(8), 3046–3052.

Betoret, N., Puente, L., Diaz, M.J., Pagan, M.J., Garcia, M.J. and Gras, M.L. (2003) Development of probiotic-enriched dried fruits by vacuum impregnation. *Journal of Food Engineering*, **56**(2–3), 273–277.

Bilbao-Sáinz, C., Andrés, A. and Fito, P. (2005) Hydration kinetics of dried apple as affected by drying conditions. *Journal of Food Engineering*, **68**(3), 369–376.

Cárcel, J.A., Benedito, J., Rosselló, C. and Mulet, A. (2007) Influence of ultrasound intensity on mass transfer in apple immersed in a sucrose solution. *Journal of Food Engineering*, **78**(2), 472–479.

Chiralt, A. and Fito, P. (2003) Transport mechanisms in osmotic dehydration: the role of the structure. *Food Science and Technology International*, **9**(3), 179–186.

Chiralt, A., Fito, P., Barat, J.M., Andrés, A., González-Martínez, C., Escriche, I. and Camacho, M.M. (2001) Use of vacuum impregnation in food salting process. *Journal of Food Engineering*, **49**(2–3), 141–151.

Contreras, C., Martín, M.E., Martínez-Navarrete, N. and Chiralt, A. (2005) Effect of vacuum impregnation and microwave application on structural changes which occurred during air-drying of apple. *LWT-Food Science and Technology*, **38**(5), 471–477.

Derossi, A., de Pilli, T. and Severini, C. (2010) Reduction in the pH of vegetables by vacuum impregnation: a study on pepper. *Journal of Food Engineering*, **99**(1), 9–15.

Fernandes, F.A.N.; Gallão, M.I. and Rodrigues, S. (2008) Effect of osmotic dehydration and ultrasound pre-treatment on cell structure: melon dehydration. *LWT-Food Science and Technology*, **41**(4), 604–610.

Fito, P. (1994) Modelling of vacuum osmotic dehydration of food. *Journal of Food Engineering*, **22**(1–4), 313–328.

Fito, P., Andrés, A., Chiralt, A. and Pardo, P. (1996) Coupling of hydrodynamic mechanism and deformation–relaxation phenomena during vacuum treatments in solid porous food–liquid systems. *Journal of Food Engineering*, **27**(3), 229–240.

Fito, P., Chiralt, A., Betoret, N., Gras, M., Cháfer, M., Martínez-Monzó, J., Andrés, A. and Vidal, D. (2001) Vacuum impregnation and osmotic dehydration in matrix engineering – Application in functional fresh food development. *Journal of Food Engineering* **49** (2–3), 175–183.

Gras, M., Vidal-Brotóns, D., Betoret, N., Chiralt, A. and Fito, P. (2002) The response of some vegetables to vacuum impregnation. *Innovative Food Science and Emerging Technologies*, **3**, 263–269.

Gras, M. L., Vidal, D., Betoret, N., Chiralt, A. and Fito, P. (2003) Calcium fortification of vegetables by vacuum impregnation: interactions with cellular matrix. *Journal of Food Engineering*, **56**(2–3), 279–284.

Guamis, B., Trujillo, A.J., Ferragut, V., Chiralt, A., Andrés, A. and Fito, P. (1997) Ripening control of Manchego type cheese salted by brine vacuum impregnation. *International Dairy Journal*, **7**(2–3), 185–192.

Guillemin, A., Degraeve, P., Guillon, F., Lahaye, M. and Saurel, R. (2006) Incorporation of pectinmethylesterase in apple tissue either by soaking or by vacuum–impregnation. *Enzyme and Microbial Technology*, **38**(5), 610–616.

Guillemin, A., Degraeve, P., Noel, C. and Saurel, R. (2008) Influence of impregnation solution viscosity and osmolarity on solute uptake during vacuum impregnation of apple cubes (var. Granny Smith). *Journal of Food Engineering*, **86**(4), 475–483.

Jeon, M. and Zhao, Y. (2005) Honey in combination with vacuum impregnation to prevent enzymatic browning of fresh-cut apples. *Journal of Food Science and Nutrition*, **56**(3), 165–176.

Khan, A.A. and Vincent, J.F.V. (1990) Anisotropy of apple parenchyma. *Journal of Science of Food and Agriculture*, **52**(4), 455–466.

Lin, D.S., Leonard, S.W., Lederer, C., Traber, M.G. and Zhao, Y. (2006) Retention of fortified vitamin E and sensory quality of fresh-cut pears by vacuum impregnation with honey. *Journal of Food Science*, **71**(7), S553–S559.

Martin-Esparza, M.E., Martinez-Navarrete, N., Chiralt, A. and Fito, P. (2006) Dielectric behavior of apple (var. Granny Smith) at different moisture contents: effect of vacuum impregnation. *Journal of Food Engineering*, **77**(1), 51–56.

Martínez-Monzó, J., Martinez-Navarrete, N., Chiralt, A. and Fito, P. (1998) Mechanical and structural changes in apple (Var. Granny Smith) due to vacuum impregnation with cryoprotectants. *Journal of Food Science*, **63**(3), 499–503.

Mavroudis, N.E., Gekas, V. and Sjöholm, I. (1998) Osmotic dehydration of apples. Shrinkage phenomena and the significance of initial structure on mass transfer rates. *Journal of Food Engineering*, **38**(1), 101–123.

Mendoza, F., Verboven, P., Ho, Q.T., Kerckhofs, G., Wevers, M. and Nicolaï, B. (2010) Multifractal properties of pore-size distribution in apple tissue using X-ray imaging. *Journal of Food Engineering*, **99**(2), 206–215.

Moreno, J., Simpson, R., Pizarro, N., Parada, K., Pinilla, N., Reyes, J. E. and Almonacid, S. (2012) Effect of ohmic heating and vacuum impregnation on the quality and microbial stability of osmotically dehydrated strawberries (cv. Camarosa). *Journal of Food Engineering*, **110**(2), 310–316.

Park, S.-I., Kodihalli, I. and Zhao, Y. (2005) Nutritional, sensory, and physicochemical properties of vitamin E- and mineral-fortified fresh-cut apples by use of vacuum impregnation. *Journal of Food Science*, **70**(9), S593–S599.

Phoon, P.Y., Galindo, F.G., Vicente, A. and Dejmek, P. (2008) Pulsed electric field in combination with vacuum impregnation with trehalose improves the freezing tolerance of spinach leaves. *Journal of Food Engineering*, **88**(1), 144–148.

Salvatori, D., Andrés, A., Chiralt, A. and Fito, P. (1998) The response of some properties of fruits to vacuum impregnation. *Journal of Food Process Engineering*, **21**(1), 59–73.

Schulze, B., Peth, S., Hubbermann, E.M and Schwarz, K. (2012) The influence of vacuum impregnation on the fortification of apple parenchyma with quercetin derivatives in combination with pore structures X-ray analysis. *Journal of Food Engineering*, **109**(3), 380–387.

Simal, S., Benedito, J., Sánchez, E.S. and Rosselló, C. (1998) Use of ultrasound to increase mass transport rates during osmotic dehydration. *Journal of Food Engineering*, **36**(3), 323–336.

Torreggiani, D. and Bertolo, G. (2001) High quality fruit and vegetable products using combined processes. In: P. Fito, A. Chiralt, J.M. Barat, W.E.L. Spiess and D. Behsnililan (eds), *Osmotic Dehydration and Vacuum Impregnation. Applications in Food Industries*. Food Preservation Technology Series. Technomic Publishers, Lancaster, Pennsylvania, USA, pp. 3–9.

Verboven, P., Kerckhofs, G., Mebatsion, H.K., Ho, Q.T., Temst, K., Wevers, M. *et al.* (2008) Three-dimensional gas exchange pathways in pome fruit characterized by synchrotron X-ray computed tomography. *Plant Physiology*, **147**(2), 518–527.

Xie, J. and Zhao, Y. (2003) Nutritional enrichment of fresh apple (Royal Gala) by vacuum impregnation. *Journal of Food Science and Nutrition*, **54**(5), 387–398.

Zhao, Y. and Xie, J. (2004) Practical applications of vacuum impregnation in fruit and vegetable processing. *Trends in Food Science and Technology*, **15**(9), 434–451.

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Refrigeration in Food Preservation and Processing

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15.1 Introduction

The aim of all food preservation methods is to slow down processes that cause deterioration of quality and wholesomeness. These may be physical processes such as loss of water by evaporation or loss of texture, chemical/biochemical processes such as enzymic degradation or microbial processes such as the growth of bacteria and moulds (Bøgh-Sørensen, 2006). Food preservation thus involves the inactivation of harmful micro-organisms and enzymes, and/or the slowing down of their growth by manipulating environmental conditions such as temperature, moisture, acidity or oxygen availability.

Traditional methods of food preservation include refrigeration (chilling and freezing), drying, heating, fermentation and additives such as salt, sugar or vinegar. Of these, refrigeration causes the least physical and chemical changes in foods and is therefore the usually preferred method for fresh, high-value foods. Since refrigeration does not eliminate microorganisms, the product remains sensitive to changes in environmental conditions, which must be constantly controlled for the best results. Thus, refrigeration technology is concerned not just with chilling and freezing, but also with cold storage, refrigerated transport, retail display and domestic storage. Refrigeration may also be applied in conjunction with other preservation methods, the

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pasteurization of milk followed by cooling and refrigerated storage being an example.

Although classified as a conventional food preservation method, the artificial refrigeration of food is very recent compared with other traditional methods such as drying, heating, salting or fermentation. In the ancient world, some food was cooled by natural refrigeration using naturally occurring ice and snow, storing in cool caves or evaporative cooling. However, systematic refrigeration was possible only with the invention of artificial refrigeration in the eighteenth century. The first known large-scale artificial refrigeration plant for food was a meat freezing plant built in Sydney (Australia) by Thomas Mort, and the first shipment of frozen meat took place in 1873 when James Harrison installed a vapour compression refrigeration system on a sailing ship to send frozen beef from Australia to the United Kingdom.

Refrigeration equipment may be divided into refrigeration providers and refrigeration applications. Refrigeration providers are machinery that create cold by moving heat energy from a medium to a higher temperature environment. In a mechanical system, this will be the compressor–evaporator–condenser circuit, which moves heat from the refrigerant to the atmosphere. Refrigeration applications are equipment that transfers heat from the product or enclosure (refrigerated cabinet or room) to the colder refrigerant. In this chapter we will only be concerned with the applications side, since refrigeration providers are the domain of mechanical engineers rather than food technologists.

The cost of refrigeration increases as the required temperature decreases. Industrially, food refrigeration temperatures may be divided into the following principal ranges:

- Moderate cooling involves bringing the food to a moderately low temperature (typically 15 to 20 °C), which can be done cheaply with cold water or evaporative cooling. In most cases this is followed by further chilling or freezing.
- Chilling involves cooling the food to a temperature not far above its freezing point, which for most fresh foods is in the range 0 to −2 °C (Miles *et al.*, 1997), usually using mechanical refrigeration. Chilling may be followed by freezing. The staging of the refrigeration process reduces energy cost, since lower temperatures are more expensive to obtain. A technique known as superchilling aims to bring the food temperature to just *below* the freezing point, the partial freezing conferring greater temperature stability. Chilling is not sufficient to stop all microbial growth, but because there is no phase change, which causes at least some loss of texture and extra moisture loss in most foods, chilled food is considered fresher than frozen food and usually fetches a higher price than the latter.
- Conventional freezing usually attempts to bring the food down to the range −12 to −40 °C, which is readily done by mechanical refrigeration.

Some bacteria and moulds may continue down to −10 °C while biochemical changes may continue at even lower temperatures, albeit at a very slow rate. Frozen meat is usually kept at −18 °C, fish in the range −18 to −30 °C, while ice cream is brought down to −35 to −40 °C for hardening and then stored below −25 °C to prevent crystal growth and texture loss. At a temperature known as glass transition, which is specific to each food, most changes become negligibly slow. Glass transition temperatures ranging from −20 to −35 °C are reported for a number of fresh fruit, vegetable, meat and fish (Reid *et al.*, 2003).

- Cryogenic freezing covers the lowest temperature ranges. The most common cryogenic refrigerants are dry ice (CO_2), with a sublimating temperature of −78.5 °C, and liquid nitrogen, with a boiling point of −196 °C. Although theoretically cryogenic freezing may ensure better retention of food quality than conventional freezing, in practice this difference is rarely noticeable.

15.2 Changes in foods during refrigeration

The prime objective of food refrigeration is to preserve food quality and safety. Product quality factors include visual appearance, texture, taste, flavour and nutritional contents. The rate of deterioration depends on the cooling rate and the final product temperature, as well as on nonthermal factors such as pre-treatment and packaging. It is not always true that the fastest cooling rate will lead to the best quality. Product safety is governed by the growth of pathogenic microorganisms. On the other hand, economics depends primarily on the refrigerating temperature: the lower the temperature, the more expensive the process.

15.2.1 Moisture movement

In air cooling and freezing, moisture loss from food is often a major consideration. When unwrapped fresh food such as meat or vegetable is exposed to cold air, the water vapour pressure at the surface of the warm food is higher than that in the cooler air, and evaporation will occur. Moisture loss may cause loss of texture, an unattractive appearance and loss of juiciness, especially in vegetables and fruit. In frozen meat, ice sublimation can cause a glassy appearance known as freezer burn.

Apart from quality aspects, moisture migration has a significant quantitative effect in the form of product mass loss. The water that is lost has the same economic value as the product it evaporates from and the loss can greatly affect profitability. Typical weight losses during meat processing, for example, amount to about 1–2% during chilling, 1% during freezing and about 0.5% per month during storage and transport, unless the product is wrapped in an

impervious film (Pham, 1987; Pham and Willix, 1984). On the other hand, a certain degree of moisture loss is sometimes necessary to improve food quality and safety. During the chilling of meat, surface drying will prevent or retard microbial growth by reducing the surface water activity.

Moisture loss during chilling and freezing is minimized by fast chilling and freezing and by wrapping the product in a low permeability film. For unwrapped products, moisture loss during storage is minimized by keeping a low temperature, high air humidity (as close to 100% relative humidity as possible) and low air velocities. Temperature fluctuation during storage should be minimized and exposure of the product to radiative heat sources such as lights or warm surfaces should be avoided.

15.2.2 Ice formation and crystal growth

Usually the water in the food does not turn into ice until it is cooled several degrees below the thermodynamic freezing point. At moderate freezing temperatures, for cellular tissues, such as meat and vegetables, ice crystals will form in the extracellular space and continue to grow as moisture gradually moves from inside the cells to the outside. The growth of crystals causes cell distortion and damage to cell membranes (Anon and Calvelo, 1980). On thawing, water is not completely re-absorbed and some is lost as drip. Faster freezing causes smaller ice crystals to form, and at very fast rates, such as with liquid nitrogen freezing, intracellular ice will form. In principle, the smaller the ice crystals, the less damage occurs and the higher the quality, although there are exceptions to this rule (Anon and Calvelo, 1980). However, to make the most of fast freezing the product should be of small size, such as peas or diced vegetables, or well agitated, such as ice cream, and in direct contact with the cold medium or cold surface. With large products such as chunks of meat, or when the product is packed in bags or cartons before freezing, most of the product will freeze slowly.

During frozen storage, water moves from small crystals towards larger ones and eventually the smaller crystals disappear. The average crystal size thus increases and the beneficial effects of fast freezing are lost (Pham and Mawson, 1997). This process is accelerated when the storage temperature fluctuates due to lack of control, especially during transport, retail display and home storage.

When changing into ice, water expands by about 9%. In some cases this may cause freeze cracking, especially during cryogenic freezing and in the freezing of large fish such as tuna (Hung and Kim, 1996).

15.2.3 Physiological changes

Chilling may cause damage to the cell membrane of fruit and vegetables, leading to other reactions that damage or kill the cells and cause loss of texture,

colour and other quality factors. Chilling injury may happen at temperatures well above freezing, especially with tropical fruit and vegetables.

Fruit and vegetables continue to respire after harvest, consuming oxygen and giving off carbon dioxide, water and heat. Respiration slows down at low temperatures and ceases completely upon freezing, which kills the cells. The design of chillers must take the heat of respiration into account. Cartons and other containers for respiring products must have slots or holes in the walls and gaps in the product stack to allow air to pass through the product and carry away the metabolic heat. For nonliving products, including all frozen products, it is better to arrange for cold air to circulate around the product stack to prevent heat gain.

15.2.4 Biochemical changes

Biochemical reactions are influential in affecting sensory qualities: texture, colour, taste and flavour. Biochemical reactions such as ripening, starch–sugar reactions in vegetables, autolysis (enzymic tissue destruction), oxidation and vitamin degradation slow down at lower temperatures and most are stopped by freezing. However, some important reactions may continue below the freezing point, such as lipid auto-oxidation, leading to rancidity in fish and meat. Some reactions, such as protein denaturation, may proceed most rapidly at temperatures just below the freezing point, due to the concentrative effect of freezing: as water turns into ice, the solution that it leaves behind becomes more concentrated in salts and other solutes. At lower temperatures the thermal effect still predominates and slows down all processes.

Some biochemical reactions are desirable and should be allowed to proceed before chilling or freezing is carried out. For example, if beef is chilled to below 15 °C immediately after slaughter it undergoes cold shortening and becomes very tough. This can be avoided by electrical stimulation followed by a period of ageing. In other cases, undesirable enzymic reactions can be stopped by blanching before freezing.

15.2.5 Microbial growth

Refrigeration aims at stopping the growth of both *pathogenic* bacteria, which are dangerous to health, and *spoilage* bacteria, which cause an unpleasant taste, smell or appearance. A food may retain acceptable smell and taste but contain dangerous bacteria or it may look or taste spoiled but is still safe to eat. Microbial growth depends on several factors: temperature, water activity (a function of moisture), pH, the presence of preservatives, gaseous atmosphere, etc. If other conditions are optimal bacterial growth may continue down to −7 °C and some yeasts and moulds down to −10 °C (Gill, 2012). Upon thawing, microorganisms go through a lag period before starting to

grow. Microorganisms normally grow only on the surface of foods, including a cut surface. Cut and especially minced or ground foods are most at risk. During freezing, the risk is normally small except for minced meat because the surface is exposed to subzero temperatures. The greatest risk occurs during thawing, when the surface is exposed to temperatures above zero for long periods. Freezing will kill a fraction of the microorganisms but this cannot be relied on as a sterilization method.

15.3 Chilling and freezing time prediction

15.3.1 The heat transfer coefficient

The heat transfer rate Q (in Watt) from an object to the surroundings can be described by Newton's cooling law:

$$Q = hA(T_s - T_a) \tag{15.1}$$

where T_s and T_a are, respectively, the object's surface temperature and the surrounding temperature in °C or K, A the object's surface area in m^2 and h is the overall heat transfer coefficient in W/m^2 K. The inverse of h measures the external resistance to heat transfer and may include factors such as the thermal resistance of a container's wall or wrapping (which hinders heat transfer) and radiation or evaporative cooling (which enhances it). Higher heat transfer coefficients result in shorter chilling and freezing times and more compact equipment, especially when the product size is small, such as with particulate or thin products. Typical heat transfer coefficient values are listed in Table 15.1. These values are only indicative, as estimates vary widely between sources and special designs or unusual operating conditions may significantly extend the values.

Table 15.1 Typical heat transfer coefficients for common equipments

Type of equipment	Heat transfer coefficients (W/m^2 K)
Tubular heat exchangers (fluids)	150–1200
Plate heat exchangers (fluids)	1000–4000
Still air chillers and freezers	5–10
Air blast chillers and freezers	15–30
Impingement chillers and freezers	50–200
Fluidized bed chillers and freezers	50–250
Immersion chillers and freezers	100–300
Plate chillers and freezers	500–1000

15.3.2 Chilling time for a well-stirred liquid

For a well-stirred liquid, the temperature T is uniform throughout ($T = T_s$) and from Newtons' cooling law we can show that

$$mc_p \frac{\mathrm{d}T}{\mathrm{d}t} = hA(T - T_a) \tag{15.2}$$

where m is the liquid's mass in kg, c_p its specific heat in J/kg K and t is the time. From this equation we can predict how the liquid's temperature changes with time under constant conditions:

$$T = T_a + (T_i - T_a) \exp\left(-\frac{hA}{mc_p} t\right) \tag{15.3}$$

where T_i is the initial product temperature. The rate of heat release Q (W) is given by

$$Q = hA(T - T_a) = hA(T_i - T_a) \exp\left(-\frac{hA}{mc_p} t\right) \tag{15.4}$$

15.3.3 Chilling time for a solid

Effect of the Biot number The temperature profile in a solid is not uniform during the cooling process, due to internal resistance to heat transfer. Heat from the centre must be conducted through layers of solid before reaching the surface and then cross over to the surroundings either directly or through layers of packaging. Processing time and heat load are influenced by the size and shape of the product. The Biot number, Bi, measures the ratio of internal to external resistance:

$$Bi = hR/k \tag{15.5}$$

where R the smallest dimension and k the thermal conductivity of the product. Internal resistance can be neglected if $Bi << 1$ (high thermal conductivity, small thickness, low heat transfer coefficient), in which case the liquid chilling solution can be used as a limiting case.

It is vital for practising engineers to be aware of the Biot number of their thermal processes. If it is much larger than 1 (i.e. if the internal resistance is controlling heat transfer), then there is little use trying to improve heat transfer, say, by reducing the amount of wrapping or increasing air velocity, and the engineer should try to reduce the dimensions of the product (if possible); the reverse applies when Bi is much smaller than 1.

Chilling time of slabs, infinite cylinders and spheres When a solid's shape is or can be approximated by one of the three basic shapes (infinite slab or wall, infinite cylinder, sphere) and the thermal properties, the heat transfer coefficient and ambient temperature are constant, analytical solutions can be obtained for the cooling rate. After an initial cooling period, the temperature at every point in the solid, as well as its mean temperature, decay exponentially (Pflug, Blaisdell and Kopelman, 1965). The centre temperature T_c and mean temperature T_m fall according to the equations

$$Y_c \equiv \frac{T_c - T_a}{T_i - T_a} = j_c \exp\left(-2.303\frac{t}{f}\right) \tag{15.6}$$

$$Y_m \equiv \frac{T_m - T_a}{T_i - T_a} = j_m \exp\left(-2.303\frac{t}{f}\right) \tag{15.7}$$

where j_c, j_m (termed the lag factors) and f (the decimal reduction time) are parameters that depend on the shape, size and thermal properties of the solid; f is the time for the residual temperature difference and heat load to decrease by a factor of 1/10 during the exponential decay period. Y_c and Y_m are the dimensionless residual temperatures or unachieved temperature changes. The exponential decay period is generally considered to start when the temperature fall is more than about 30% of the maximum that can be achieved. The analytical formulas for the f and j factors are rather complicated, but Figures 15.1 to 15.3 can be used to calculate these parameters. In these figures,

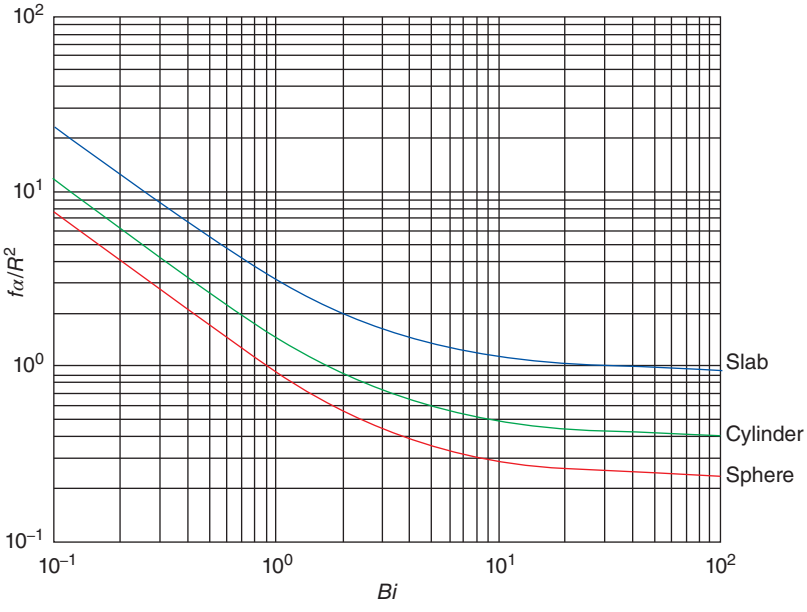

Figure 15.1 Plot of $f\alpha/R^2$ against Biot number (Bi)

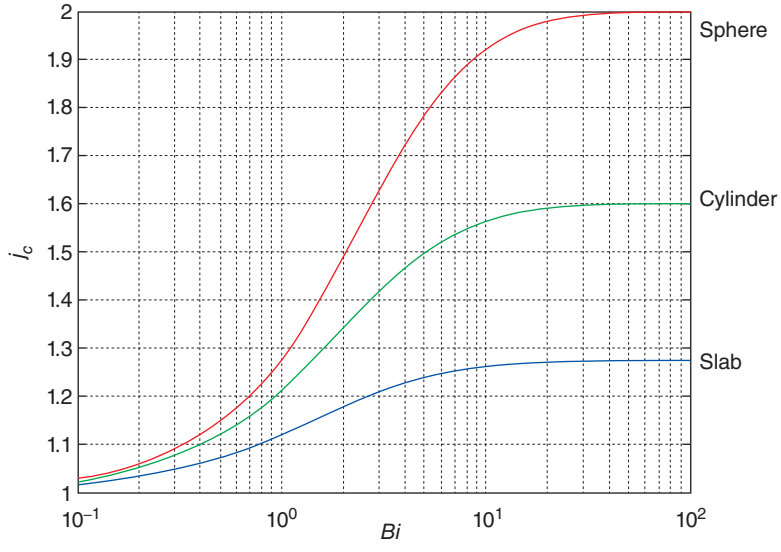

Figure 15.2 Plot of j_c against Biot number (Bi)

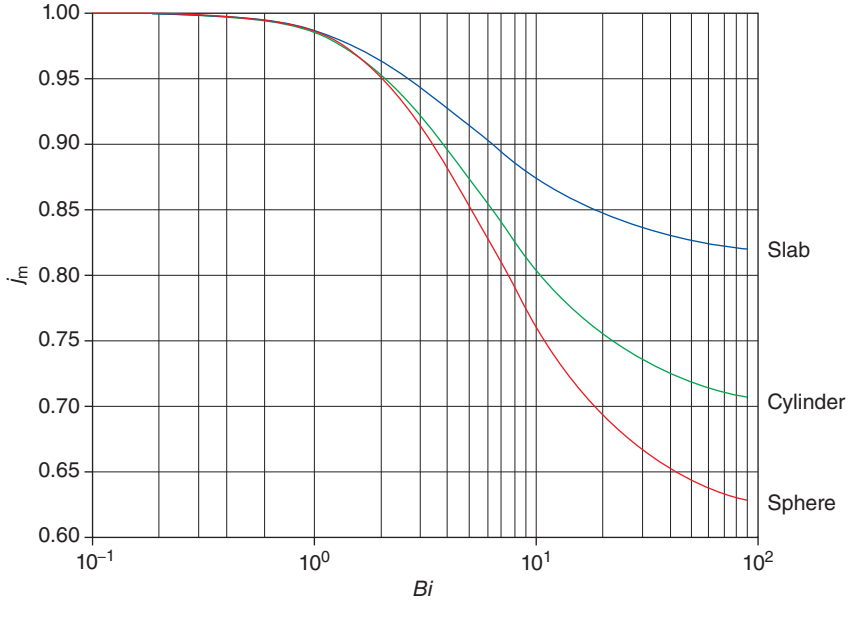

Figure 15.3 Plot of j_m against Biot number (Bi)

α is the thermal diffusivity in m^2/s, defined by $\alpha = k/\rho c_p$, where ρ is the density in kg/m^3.

For those who prefer algebraic expressions to graphs, Ramaswamy, Lo and Tung (1982) present the following approximate equations for $Bi > 0.03$ (for $Bi \leq 0.03$ the liquid chilling equations can be used). In these equations ϕ (the dimensionless decay time) is defined by $\phi \equiv f\alpha/2.303R^2$.

- For slabs:

$$1/\phi = 2.0738Bi/(Bi+2) + 0.2795\arctan(Bi/3) - 0.0291\arctan(5Bi) + 0.001171 \quad (15.8)$$

$$j_c = 0.1138\arctan(Bi) + 0.1111\arctan(Bi/3) - 0.05142\arctan(Bi/7) + 1.0016 \quad (15.9)$$

$$j_m = \frac{\phi^{-1/2}}{\sin\phi^{-1/2}} j_c \quad (15.10)$$

- For cylinders:

$$1/\phi = 4.1093\ Bi/(Bi+2) + 1.2365\arctan(Bi/3) - 0.1641\arctan(2Bi) - 0.007762 \quad (15.11)$$

$$j_c = 0.4411\arctan(Bi/2) + 0.007242\arctan(11Bi) - 0.1021Bi/(Bi+11) + 0.9984 \quad (15.12)$$

$$j_m = (1 - 0.125\phi^{-1} + 5.208\times10^{-3}\phi^{-2} - 1.085\times10^{-4}\phi^{-3} + 1.351\times10^{-6}\phi^{-4})\, j_c \quad (15.13)$$

- For spheres:

$$1/\phi = 4.0704\ Bi/(Bi+2) + 3.5560\arctan(Bi/3) + 0.1781\arctan(Bi/8) - 0.04036\arctan(7Bi) + 0.002262 \quad (15.14)$$

$$j_c = 0.4564\ Bi/(Bi+2) - 0.98978\ Bi/(Bi+4) + 0.03884\arctan(Bi/2) + 0.9370\arctan(Bi/3) + 0.9992 \quad (15.15)$$

$$j_m = 3\phi(\phi^{-1/2}\sin\phi^{-1/2} - \cos\phi^{-1/2})j_c \quad (15.16)$$

At very high Biot numbers ($Bi > 100$) the following asymptotic values are reached:

- For slabs:

$$\phi = 0.4053,\ \ j_c = 1.273,\ \ j_m = 0.811 \quad (15.17)$$

- For cylinders:

$$\phi = 0.1728, \quad j_c = 1.601, \quad j_m = 0.691 \tag{15.18}$$

- For spheres:

$$\phi = 0.1012, \quad j_c = 2.000, \quad j_m = 0.607 \tag{15.19}$$

Chilling time of other shapes For more complex regular shapes (rectangular prisms, bricks, spheres, infinite and finite cylinders, ellipses or ellipsoids), an empirical shape factor can be applied to the regular-shape solution (Pham, 2001; Lin *et al.*, 1996a, 1996b). Solid foods with irregular shapes can be approximated by the nearest regular shape and the same methodology is suitable.

15.3.4 Freezing time

The freezing of a food item can be divided into three distinct periods (see Figure 15.4):

1. A pre-cooling period, where cooling takes place without a phase change.
2. A phase change period, which starts when ice starts to form at the surface. Because of supercooling, the first ice crystals usually do not form until the surface temperature is a few degrees below the initial freezing point T_f. When ice appears, the temperature will almost immediately rise back to near T_f due to latent heat release. The ice front gradually advances into the product until it reaches the thermal centre and the whole food can be considered frozen.
3. A post-cooling period, during which the frozen food continues to cool until it reaches the equilibrium temperature.

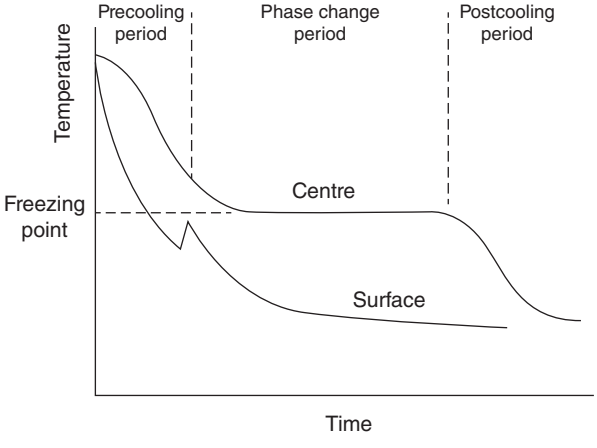

Figure 15.4 Typical temperature history during freezing

For practical purposes the freezing process is considered completed when a specified centre temperature has been reached, such as −18 °C. The latent heat of freezing is usually much greater than the sensible heat of cooling and therefore the phase change period (period 2) takes up the most time. Note that in foodstuffs, the latent heat of freezing is not released at a sharp temperature but over a range of temperatures, because as some water turns into ice the remaining unfrozen solution becomes more and more concentrated in solutes and the freezing point falls. This is shown in a graph of apparent specific heat (which includes the latent heat of freezing) against temperature (Figure 15.5).

Freezing time of slabs, infinite cylinders and spheres If the pre-cooling and post-cooling periods can be ignored, Plank's equation (Plank, 1913) can be used to predict the freezing time. For the three basic shapes the Plank equations can be written in the following common form:

$$t_{Plank} = \frac{\rho L_f R}{E_{Freeze} h (T_f - T_a)} \left(1 + \frac{Bi_f}{2}\right) \tag{15.20}$$

where E_{Freeze} (the shape factor or equivalent heat transfer dimensionality for freezing) is 1 for the slab, 2 for the infinite cylinder and 3 for the sphere, and Bi_f is calculated based on the frozen thermal conductivity. To take into account the pre-cooling and post-cooling times, Pham (1986) proposed the following equation:

$$t_f = \frac{R}{E_{Freeze} h} \left(\frac{\Delta H_1}{\Delta T_1} + \frac{\Delta H_2}{\Delta T_2}\right) \left(1 + \frac{Bi_f}{2}\right) \tag{15.21}$$

where ΔH_1 and ΔT_1 are the specific enthalpy change and temperature difference for the pre-cooling period and ΔH_2 and ΔT_2 those for the combined

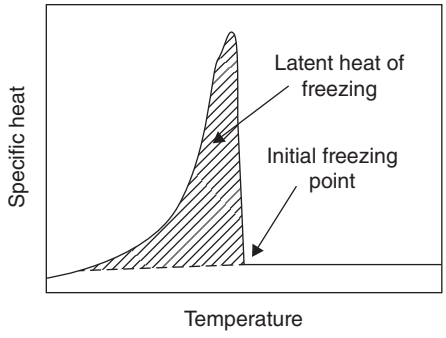

Figure 15.5 Typical plot of effective specific heat (including latent heat effect) versus temperature of water rich foods

freezing–post-cooling period, calculated from:

$$\Delta H_1 = \rho_u c_u (T_i - T_{fm}) \tag{15.22}$$

$$\Delta T_1 = (T_i + T_{fm})/2 - T_a \tag{15.23}$$

$$\Delta H_2 = \rho_f \left[L_f + c_f (T_{fm} - T_c)\right] \tag{15.24}$$

$$\Delta T_2 = T_{fm} - T_a \tag{15.25}$$

L_f is the latent heat of freezing in J/kg, ρ_u and c_u are the density (kg/m^3) and specific heat (J/kg K) of the unfrozen food, ρ_f and c_f those of frozen food, T_c is the final centre temperature and T_{fm} is termed the 'mean freezing temperature'. The following empirical equation is suggested for most water-rich biological materials (all temperatures are in °C):

$$T_{fm} = 1.8 + 0.263 T_c + 0.105 T_a \tag{15.26}$$

Freezing time of other shapes For infinite or very long rectangular rods, bricks and finite cylinders, the empirical method by Cleland, Cleland and Earle (1987) can be used:

$$E_{Freeze} = G_1 + G_2 E_1 + G_3 E_2 \tag{15.27}$$

where G_1, G_2 and G_3 are constants that depend on the type of shape, as listed in Table 15.2, while E_1 and E_2 are empirical functions of Bi:

$$E_1 = X(2.32\beta_1^{-1.77})\frac{1}{\beta_1} + \frac{1 - X(2.32\beta_1^{-1.77})}{\beta_1^{1.47}}\frac{0.73}{\beta_1^{2.50}} \tag{15.28}$$

$$E_2 = X(2.32\beta_2^{-1.77})\frac{1}{\beta_2} + \frac{1 - X(2.32\beta_2^{-1.77})}{\beta_2^{1.47}}\frac{0.50}{\beta_2^{3.69}} \tag{15.29}$$

where $X(x)$ is a function defined by

$$X(x) = x/(Bi_f^{1.34} + x) \tag{15.30}$$

Table 15.2 Geometric constants for freezing E factors

Shape	G_1	G_2	G_3
Finite cylinder, height < diameter	1	2	0
Finite cylinder, diameter < height	2	0	1
Rectangular rod	1	1	0
Rectangular brick	1	1	1

For elliptical cylinders, Hossain, Cleland and Cleland (1992) propose the following approximate expression:

$$E_{freeze} = 1 + \frac{1 + 2/Bi_f}{\beta_1{}^2 + 2\beta_1/Bi_f} \tag{15.31}$$

For three-dimensional ellipsoids, the same authors suggested the equation

$$E_{freeze} = 1 + \frac{1 + 2/Bi_f}{\beta_1{}^2 + 2\beta_1/Bi_f} + \frac{1 + 2/Bi_f}{\beta_2{}^2 + 2\beta_2/Bi_f} \tag{15.32}$$

Solid foods with irregular shapes must be approximated by the nearest regular shape, and then one of the previous relationships can be applied.

15.4 Refrigeration equipment

15.4.1 Equipment for the refrigeration of liquids

Liquid foods and drinks are cooled and heated in heat exchangers. Plate heat exchangers are the most popular types in the food industry, due to their compactness, high heat transfer coefficient (Table 15.1) and ease of cleaning (the plates can be quickly unclamped and dismantled for cleaning). The high heat transfer coefficient is due to the liquid being split into thin films flowing through narrow gaps between the plates, whose corrugations enhance turbulence. Tubular heat exchangers (jacketed tubes) are less compact and efficient than plate heat exchangers. Liquid food and drinks can also be cooled in vessels with jackets and/or immersed cooling coils, but heat transfer rates in these are lower and therefore these equipments tend to be used only for the storage of pre-chilled liquids.

Viscous liquids such as yoghurt, sauces and ice cream can be chilled or partially frozen in scraped surface heat exchangers, which are tubular heat exchangers equipped with internal rotating scraper blades which keep the liquid agitated and continually bring new liquid into contact with the cold walls.

15.4.2 Equipment for the refrigeration of bulky foods

Bulky foods are solid foods in large chunks or blocks, such as meat carcasses, meat cuts, blocks of butter, cartoned foods or containers of liquid foods and drinks. Due to their large size, the internal resistance to heat transfer is appreciable, and therefore there is little benefit in providing very high surface heat transfer coefficients. Figure 15.6 shows a few common types of chillers and freezers for bulky foods.

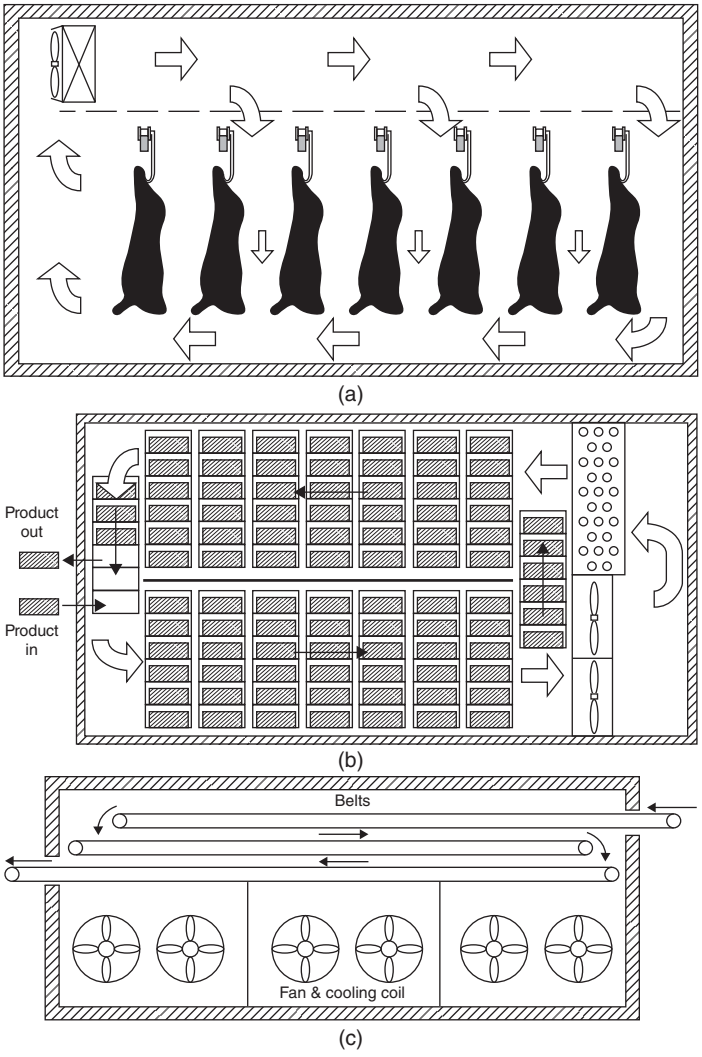

Figure 15.6 Types of chillers and freezers (a) lamb air blast chiller with slotted air distributor, (b) carton tunnel freezer with moving racks, (c) belt freezer, (d) spiral freezer, and (e) carton plate freezer. Black arrows: product flow, white arrows: airflow.

Still air chillers and freezers Food may be chilled or frozen in still or semi-still air. This happens in domestic refrigerators and freezers, but may also be used in small commercial installations where the food is refrigerated in cold rooms. Heat transfer is due mainly to natural convection and radiation. If air is circulated around the room, the main aim is to ensure an even temperature distribution rather than to enhance heat transfer. Heat transfer coefficients are low (Table 15.1).

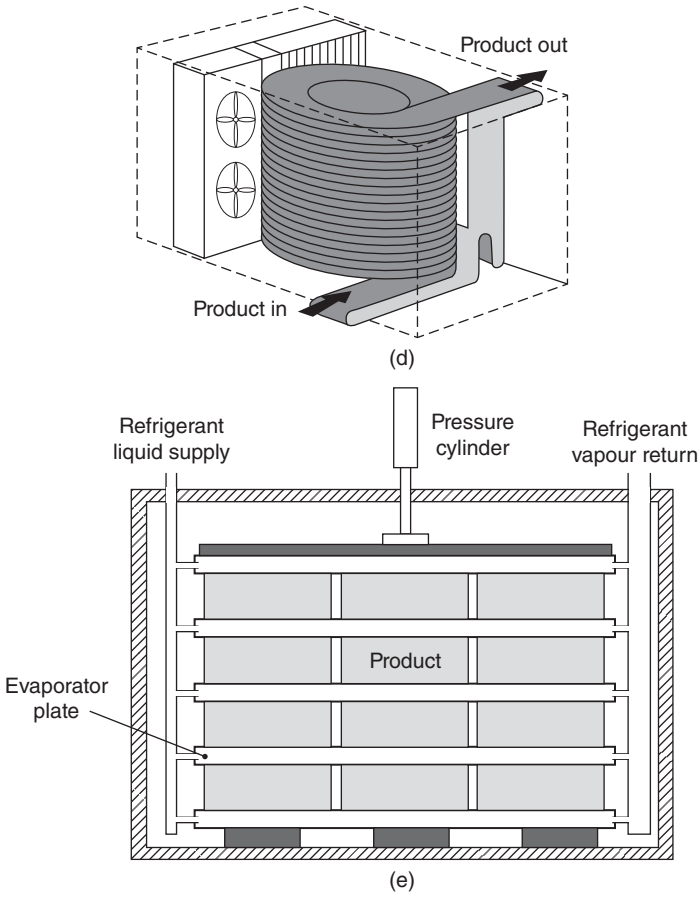

Figure 15.6 (*continued*)

Air blast chillers and freezers Air is blown at a high speed (typically 1.0 to 5.0 m/s) at and around the product, providing higher heat transfer coefficients than in still air (Table 15.1). Air blast chillers and freezers differ in the way the product is supported and handled. In batch chillers and freezers, the product may be hung on rails, as for meat carcasses, or stacked on pallets or racks. In continuous chillers and freezers, the product may be circulated on moving hooks, trolleys, racks, linear belt conveyors, spiral belt conveyors or other conveying systems.

The efficiency of air blast chillers and freezers depend on the airflow distribution around the product, particularly in batch freezers (with continuous freezers the pieces of product traverse different zones and are exposed to the same treatment). For best results, the air must be evenly distributed and free of bypasses and dead zones. This is ensured by careful geometric design of the product space, using air deflectors, keeping the product off the floors and

walls, and leaving adequate spaces for airflow between the product items to avoid unexposed surfaces (Figure 15.7).

Contact chillers and freezers In contact chillers and freezers, one or both surfaces of a flat product are in contact with a cold surface. In contact belt freezers, often used for freezing fish fillets and meat patties, this surface is a metal conveying belt. In mould chillers and freezers, the product is contained in a refrigerated metal mould. In plate chillers and freezers, the product is pressed between two refrigerated metal plates. If the food is soft and has a wet surface and sufficient pressure is applied, there will be good contact between the food and metal so that high heat transfer coefficients can be obtained (Table 15.1); the precise value is rarely of importance since the internal resistance and that of the wrapping, if any, are usually the controlling factors. Plate and mould chillers and freezers must usually be operated manually, although very large automated plate freezers are available for cartoned meat (Visser, 1986).

Immersion chillers and freezers Plate freezers require that the product has a regular geometry with parallel surfaces. For irregular-shaped products, such as fish and poultry, fast freezing can be achieved in immersion chillers and freezers, where the solid is immersed in a cold liquid. Below freezing point a brine, a sugar solution, an alcohol solution (glycol) or another nontoxic liquid mixture may be used. A recent immersion chilling medium is ice slurry, a mixture of water or aqueous solution and ice particles (usually in the range 0.1 to 1.0 mm diameter); because ice stores latent heat, the temperature is held stable and uniform at the freezing point, while the heat released or absorbed per unit volume is much higher than that of liquid water. Food freezing can be achieved by adding a solute to the ice slurry, which depresses the freezing point (Fikiin *et al.*, 2003). To avoid contamination and absorption of liquid,

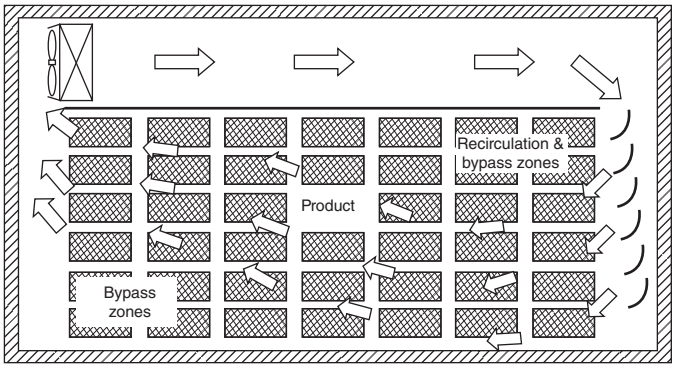

Figure 15.7 Airflow distribution in a freezer – possible problems. Arrows indicate local airflow direction.

the product may be wrapped tightly in plastic. The liquid may be circulated in a tank or sprayed on the food to be frozen.

15.4.3 Equipment for the refrigeration of thin or particulate solids

With thin products such as meat patties and fish fillets or particulate products such as peas, corn kernels and diced vegetables, the internal resistance heat transfer is small and the rate of freezing depends mainly on the surface heat transfer coefficient. Also, these products have a large surface area to volume ratio and thus tend to be susceptible to moisture loss. Thus it is necessary or advantageous to chill or freeze these products as quickly as possible, using high heat transfer coefficients and low temperatures. Any of the freezer types for bulky solids can also be used for thin or particulate solids, but an air blast belt freezer (tunnel or spiral), direct contact freezers and cryogenic freezers are particularly suitable. When using air blast freezing, the product should be spread out to minimize thickness. Two types of freezers have been especially designed for particulate solids: fluidized bed freezers and impingement freezers. The latter is also particularly suitable for thin products.

Fluidized bed chillers and freezers In fluidized bed freezers (also known as IQF for individually quick frozen), the product is conveyed on a perforated belt or along a shallow trough with a perforated bottom. Cold air is blown through the porous bottom and flows upwards through the product. Usually the freezer is divided into two zones: in the first, the particles are exposed to fast flowing, very cold air to crust-freeze in a fluidized state and avoid sticking due to freezing moisture; in the second, the fluid may be conveyed in a thicker layer to complete the freezing process economically.

Impingement chillers and freezers Cold air is forced through small orifices or nozzles on to the product, imparting high air velocity and turbulence and increasing the heat transfer coefficient. With particulate products the impinging air may also cause fluidization as it is reflected from the bottom of the bed. With both fluidized bed and impingement freezers, heat transfer coefficient values intermediate between those of air blast and immersion freezing are obtained (Table 15.1).

15.4.4 Vacuum cooling

Vacuum cooling is often used for vegetables, especially leafy vegetables such as lettuce (Sun and Wang, 2001). The product is put in a vacuum chamber. The low vapour pressure promotes water evaporation, which absorbs latent heat and lowers the product's temperature. In leafy or porous products, such

as lettuce and mushroom, this water loss takes place throughout the product, leading to much faster cooling of the core than more conventional cooling methods, in which the surface may cool quickly but the core remains warm for much longer times. To prevent excessive dehydration, the product may be pre-wetted before cooling.

15.4.5 Cryogenic freezers

Cryogenic freezing is carried out with carbon dioxide or liquid nitrogen. Carbon dioxide is supplied as a pressurized liquid or as dry ice. Upon release of pressure, part of the liquid instantly vaporizes while the remainder solidifies into dry ice, which sublimates at −78.5 °C. Liquid nitrogen vaporizes at −196 °C. The cryogen is sprayed on the food or into the air above it in a controlled manner. The heat transfer coefficient is intermediate between air blast freezing and immersion freezing, depending on the amount of direct contact between the cryogen liquid or solid and the food. From the product point of view, the main advantage of a cryogenic freezer is a lower temperature, leading to faster freezing, especially with liquid nitrogen, and hence a better product quality with less moisture loss. Often cryogenic freezing is used to freeze only the outside layer of the product (crust freeze) to harden the surface and minimize the loss of moisture, before the freezing is completed in a second stage at more moderate temperatures.

15.5 Refrigerated storage and transport

The design of refrigerated storage and transport equipments and systems must aim to maintain the required temperature, minimize temperature fluctuations, minimize radiation heat sources (warm surfaces) and, if there are unwrapped products, minimize moisture loss by maintaining high moisture and low air movement.

15.5.1 Design and operational factors during storage and transport

Initial temperature of product The product must be fully refrigerated prior to entering storage. On exit from a chiller or freezer, the product temperature may be nonuniform, the outside being colder than the centre, but the mean temperature should ideally be equal to the intended storage temperature and certainly not higher. Because cold stores are designed to maintain constant temperature by removing heat from nonproduct sources (conduction through walls, lights, air infiltration, etc.) rather than to cool down the product, if a warm product enters storage it will take a long time to cool down and its shelf-life will be affected. Furthermore, the warmer new product will also

heat up other products around it and cause temperature fluctuations and spatial variations, which are harmful to product quality and can increase moisture loss.

Air circulation Adequate air circulation must be provided to remove heat from any warm surface, such as ceiling, walls, floors and light sources, and to ensure temperature uniformity. A certain amount of air circulation should also be maintained around the product, to remove respiration heat, if any, as well as to provide cooling for a product that enters the store at a higher than storage temperature (although this should be avoided). However, when there is an unwrapped product, high air velocity must be avoided to minimize moisture loss.

Spatial temperature variations Spatial temperature variations exist in all cold stores, although well-designed and operated stores will be able to keep these to a minimum. Warm spots are near doors that open frequently, near the floor, ceiling and walls, and under the lights. A vertical temperature gradient is commonly found since warm air rises. Large subzero cold stores usually have underfloor heating to prevent the ground freezing ('frost heave') so heat conducted through the floor can be a particular problem if air circulation is poor. Products should never be stacked directly on the floor or against the walls as this impedes air circulation and heat gained through the enclosure cannot be carried away. An unwrapped water-rich product is particularly sensitive to moisture loss when there are spatial temperature variations, because moisture tends to travel from warm regions towards colder regions, causing a low relative humidity in warmer areas. Overloading or a poor loading pattern will impede the airflow and aggravate temperature nonuniformity.

Spatial variations are more serious in transportation than in cold storage, especially in road and rail transportation, due to the thinner insulation of vehicles, ageing or damaged insulation and door seals, large door area per unit volume, the long narrow shape of the container or vehicle, and extreme external airflow conditions due to movement. Refrigerated containers and trucks are usually equipped with an evaporator–fan unit at one end and a loading door at the other. Due to the lack of airflow and possible air infiltration through the door, products at the door end may be ten or more degrees warmer than those near the air delivery (Smale, Moureh and Cortella, 2006). Transport containers and vehicles are also vulnerable to solar radiation.

Refrigerated containers usually have T-bar floors to allow air circulation under the product. Air is delivered through an opening to the slotted floor, travels along the bottom towards the door end and back along the ceiling (Figure 15.8). In refrigerated road vehicles the cold air may be delivered along the ceiling. Refrigerated products should not be stacked directly on the floor

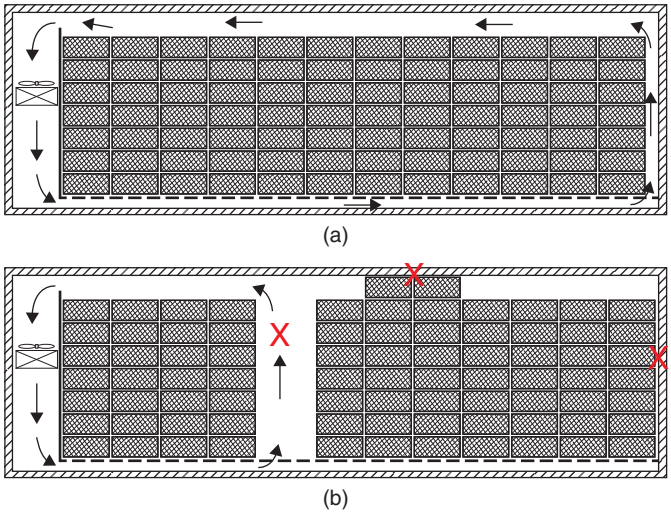

Figure 15.8 Airflow pattern in refrigerated container (a) desirable airflow pattern going around product, and (b) bad airflow pattern due to poor stacking (gap in product stack causing bypass, product too close to the door and ceiling causing blockage).

of a vehicle without a slotted floor, as they will pick up heat conducted through the floor without being cooled by the airflow. It is also desirable to ensure that the product does not rest directly against the walls, for the same reason. The product should be stowed as a solid block to force air to travel around the stack to the far end of the container or vehicle. Any gap in the product stack will allow air to bypass the rest of the container.

Mixed loads For short road trips, products at different temperatures are often loaded on the same vehicle. For single-compartment vehicles this is not good practice as the products will deviate from their optimal storage conditions. For example, tropical fruit that are sensitive to chilling injury may be damaged by a nearby superchilled product, or an ethylene-sensitive product may be affected by an ethylene-producing product. However, mixed loads can be safely carried in multicompartment vehicles, each compartment being controlled at a different temperature.

Temperature fluctuations Temperature fluctuations during cold storage are usually smaller than during transport, but can still have important effects due to the long storage time. Fluctuations are caused mainly by door openings. They can be reduced by having suitable door protections: plastic strip curtains, air curtains, air locks or refrigerated loading areas outside the door.

Well-designed plastic strip curtains can reduce air infiltration by about 93% and air curtains by about 79% (Pham and Oliver, 1983), but the main doors should still be closed whenever a door is not in use. Doors that open frequently should not be situated at opposite ends of a store as a draft will be created. Some cold stores have openings above the door level, for example to accommodate a conveyor. If not suitably protected, strong natural convection currents may be set up. Small cold stores are vulnerable to temperature fluctuations during defrosting periods.

Severe temperature fluctuations may happen during loading and unloading of vehicles if proper precautions are not taken. Ideally the refrigerated vehicle or container should connect directly with the cold store via an opening with a tight seal, such as inflatable flaps. For large installations the loading and unloading may be carried out in a refrigerated loading area enclosing the store doors. Before loading a refrigerated product into a truck or container, the latter should be pre-cooled to the desired transporting temperature, except when loading is carried out in ambient conditions, in which case condensation on the cold walls and ceiling may damage the cargo. The loading time must be minimized. On long trips, containers are particularly vulnerable to heat and solar radiation when travelling through tropical regions.

Chilled products are more vulnerable to temperature fluctuations than frozen products. Due to the latent heat of freezing, the latter rarely reach thawing temperature. Also, chilled products are more susceptible to changes in temperatures than frozen products as all biological and biochemical processes are faster above freezing. Superchilled products, with a significant fraction of moisture frozen and kept just below the freezing point, have good thermal stability since it takes an appreciable amount of heat to melt the ice and raise their temperature above the freezing point.

Air humidity For unwrapped water-rich foods, such as vegetables, fruits and meats, moisture loss is a major factor and it is important to maintain high relative humidity in the cold store. Any heat source except moisture-carrying sources (air infiltration) will tend to decrease relative humidity. Thus high relative humidity can be ensured by having good insulation, since poorly insulated walls, ceilings and floors can get warm and act as giant radiators; by ensuring that the cooling coils have large heat transfer surface areas so that they can operate at small temperature differences, since large temperature differences increase moisture condensation on the coils; and by using high efficiency lighting and turning off the lights when not in use.

15.5.2 Equipment for refrigerated food transport

Intermodal containers Intermodal refrigerated containers can be used for land as well as sea transport. The refrigeration unit is at one end of the

container and delivers cold air through a slot near the bottom (Figure 15.8). The air travels along a slotted (T-bar) floor, turns around at the door end and travels back along the ceiling to the refrigeration unit. The product must be stacked properly, avoiding gaps that would allow air to bypass the furthest part of the stow, but with sufficient space below the ceiling and gap next to the door for air return, and a small gap between the product and the walls so that the conducted heat can be carried away by cold air (Figure 15.8).

Container insulation and door seals can easily get damaged and should be regularly inspected and repaired if necessary. In addition, the insulation ages steadily, losing about 5% of their insulation value per year (Estrada-Flores, 2012).

Noncontainer sea, road and rail transport In addition to intermodal refrigerated containers, which may be used across sea, road and rail transport, there are also equipment specifically designed for each mode of transport.

(a) Sea transport Reefer (refrigerated) ships may be of two types: container ships and ships with refrigerated cargo holds. Container ships may be dedicated to refrigerated products or carry refrigerated containers along with ordinary containers. Ships with refrigerated holds may be loaded through side doors on the ship's hull or from the top using cranes. During the loading of conventional ships, the chilled or frozen product may be subjected to large temperature rises if the process is not achieved quickly and weather conditions are adverse (high wind, hot and sunny weather). Typically pallets of products are unloaded from trucks, left on the dock for some time, then loaded into the open hold with cranes, a process that may take several hours during which no refrigeration is applied.

(b) Road transport For shorter distances refrigerated panel vans may be used. Long distance road transport relies on refrigerated trucks, semi-trailers and trailers (hereafter all referred to as 'vehicles'). The ATP Agreement (2011) classifies transport vehicles for refrigerated foods into three types: insulated equipment, refrigerated equipment and mechanically refrigerated equipment. Refrigerated equipment uses a nonmechanical source of cold, such as eutectics or dry ice. Eutectics (phase change materials) may be stored in hermetic compartments or beams under the roof and must be frozen before loading, while dry ice is loaded into open compartments under the roof. For transportation at −10 °C or below, the conductance (K-coefficient) of the equipment must be equal to or less than 0.40 W/m^2 K. Multicompartment vehicles may be used for transporting products at different temperatures.

High heat loads and temperature rises occur during loading and unloading of refrigerated vehicles. Ideally the loading/unloading should be carried out in a refrigerated environment or through sealed doors to prevent air ingress.

(c) Rail transport Refrigerated foods may be transported by rail using refrigerated wagons or mechanically refrigerated wagons, as defined by the ATP (see the previous section).

Air transport Refrigerated air transport is increasingly used for high-value products. It uses special insulated containers that are smaller than ship containers and may be cooled by phase change materials, dry ice or battery-powered mechanical refrigeration units. Although air transport is very fast, the product may spend hours waiting for loading, particularly due to security-related delays. Careful scheduling will ensure that the waiting periods are kept to a minimum and precautions must be taken so that the temperature does not rise excessively during those periods.

15.5.3 Controlled atmosphere storage and transport

Controlled atmosphere (CA) is widely used to extend the post-harvest time of horticultural products (Thompson, 2010). To slow down physiological changes, the oxygen, CO_2 and moisture contents around the product are carefully monitored and controlled. This together with refrigeration will slow down respiration, delay ripening and ethylene production, retard mould growth and decay, and reduce physiological disorders such as chilling injury. The precise optimal compositions are product-specific.

As long as they are gas-tight, modern refrigerated stores and transport equipment can be converted to a controlled atmosphere by fitting gas generation (mainly nitrogen generators and CO_2 absorbers) and control equipment. The oxygen content is reduced by flushing with nitrogen, which can be generated by pressure swing absorption or a membrane system. CO_2 is produced by respiration or can be added from bottles and can be reduced by controlled absorption. Ethylene is controlled by absorption. Water vapour can be added as necessary to maintain a constant relative humidity.

For sea transport it is estimated that 20–30% of cargo on ships with refrigerated holds is transported under CA conditions. The percentage of containers operating under CA is smaller, around 4–4.5% in 2000 (German Institute of Refrigeration, 2012). CA containers may use passive or active systems. In the former, the product develops its own atmosphere by consuming oxygen and producing carbon dioxide. Once the required concentration of oxygen is achieved it is controlled by a valve to the atmosphere. Carbon dioxide is removed with lime and ethylene with potassium permanganate. In active systems, the container is flushed with nitrogen. CO_2 is produced by the product metabolism but can also be added from bottles.

15.6 Recent developments in food refrigeration

15.6.1 Developments in techniques and equipment

The following technologies are mostly still in the research stage.

High pressure freezing and thawing An increase in pressure will cause the freezing point of water to fall. The minimum freezing point that can be obtained for pure water is −22 °C at a pressure of 210 MPa. In pressure shift freezing, the product is cooled under high pressure to several degrees below its normal freezing point. Pressure is then suddenly released, causing spontaneous ice nucleation throughout the product. This results in a large number of small ice crystals, which minimize ice crystal-induced damage to the texture and cellular structure of the food (Le Bail *et al.*, 2002; Kanda, Aoki and Kosugi, 1992).

In pressure-assisted thawing, the whole thawing process is carried out at high pressure. The product will remain at the (lowered) freezing point until it is thawed, resulting in a larger temperature difference between the product and the thawing medium for a given temperature of the latter, and hence a shorter thawing time. The main drawback of high pressure processes is the cost of equipment.

Ultrasound-assisted freezing Freezing is carried out in a liquid medium subjected periodically to ultrasound (Li and Sun, 2002). By tuning the power and timing of ultrasound, it can be used to accelerate freezing, by increasing the rate of heat transfer at the surface, or to reduce crystal size during food freezing. To get the latter effect, the food is supercooled then nucleation is initiated by a short pulse of ultrasound.

Dehydrofreezing In dehydrofreezing the food is first dehydrated by air drying or osmotic dehydration (osmodehydrofreezing) and then frozen (Fikiin *et al.*, 2003). Osmodehydrofreezing involves immersing the food in a cold concentrated aqueous solution. For fruit, as water is sucked out, some solute will diffuse into the food, changing its taste. For fruit, sucrose is the most popular solute, although glucose, fructose, lactose, maltodextrin and corn syrup can also be used. For vegetables, sodium chloride can be used. Typically about 70% of the water is removed. The reduced amount of water decreases the number of ice crystals, their size (due to increased viscosity, which hinders diffusion) and the freezing expansion, and so may reduce tissue damage during freezing, especially in fragile foods such as strawberries. At least 50% of

the water must be removed to give improved texture after freezing and thawing. The increased solute concentration due to both water loss and solute gain depresses the freezing point and increases the glass transition temperature, leading to better stability, especially if cryoprotectants and cryostabilizers are added to the solution to prevent nucleation. Pigment, vitamin and aroma retention are improved. The product will taste different and may be used as an ingredient, for example for yogurt. The freezing time is shortened because there is less water to freeze. In addition the cost of packaging and transport is reduced due to reduced weight.

Freezing in an electrostatic field The application of an electrostatic field of several kilovolts during freezing causes nucleation to occur at a higher temperature. It decreases the degree of supercooling and may cause a decrease in ice crystal size (Orlowska, Havet and Le Bail, 2009). This research is promising but is still in its early stages.

Radiofrequency-assisted freezing In this technique, low voltage (2 kV) RF pulses are applied during cryogenic freezing (Anese *et al.*, 2012). The thawing losses of RF cryogenic frozen meat are much lower than that observed during thawing of air and cryogenic frozen meat, and the cellular structure is less disrupted when RF is applied, due to smaller ice crystals. This can be attributed to the ability of RF to depress the freezing point, thus producing more nucleation sites.

15.6.2 Modelling and simulation of refrigeration processes

Recent advances in computer technology and computation methods have allowed refrigeration processes to be better understood, leading to more efficient design and operation. Heat and mass transfer in solids can now be modelled with finite element method (FEM) software such as Ansys, Comsol or Abaqus. Processes involving fluid flow such as air blast freezing and immersion freezing can be modelled with computational fluid dynamics (CFD) software such as Fluent and CFX. Such software enables accurate prediction of product temperatures and heat loads during chilling and freezing, the design and optimization of airflow in chillers and freezers, cold stores, refrigerated vehicles, retail display cases, etc. (Sun, 2007). From the temperature and moisture histories, the evolution of food quality factor and microbial numbers can also be predicted by computer software such as those of the University of Nebraska–Lincoln (2012) and Meat and Livestock Australia (2012).

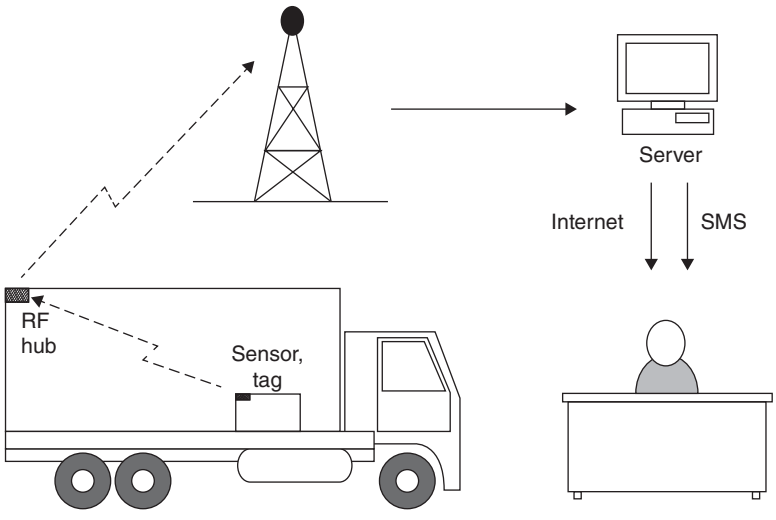

Figure 15.9 RFID system for monitoring cargo and air temperatures in a refrigerated transport system

15.6.3 Cold chain monitoring

Food quality and the health and safety of consumers depend crucially on maintaining temperature and other environmental factors within required values during storage and transport, and minimizing fluctuations. Recent advances in sensing technology, miniaturization and remote communications have resulted in small but powerful and accurate temperature and humidity sensors and loggers that provide continuous recordings of product and environment temperatures during various stages, so that shortcomings in the cold chain can be identified and remedied. The environmental data are recorded to memory and, using RFID (radio frequency identification) technology, can also be transmitted and monitored continuously, together with data on geographical location, using mobile wireless systems and the Internet, allowing corrective actions to be taken in real time when necessary (International Institute of Refrigeration, 2008; Ruiz-Garcia *et al.*, 2009) (Figure 15.9). According to the IIR, the main limitations of RFID at the moment are high cost, difficulties in calculating the rate of return and the need to improve reliability and accuracy.

15.7 Conclusions

Refrigeration is likely to remain the principal preservation method for high-quality, nutritious foods that retain all or most of the characteristics

of freshness. With the growth of the urban middle class in industrializing countries, demand for refrigerated foods will continue to increase rapidly for the foreseeable future. Continued research in refrigeration techniques and the application of modern computer and communication technologies to the modelling, monitoring and control of the cold chain will produce better and safer foods while reducing energy consumption and environmental impact.

References

Anese, M., Panozzo, A., Beraldo, P., Foschia, M. and Nicoli, M.C. (2012,) Effect of radiofrequency assisted freezing on meat microstructure and quality. *Food Research International*, **46**, 50–54.

Anon, M.C. and Calvelo, A. (1980) Freezing rate effect on the drip loss of frozen beef. *Meat Science*, **4**, 1–14.

ATP Agreement (2011) Agreement on the international carriage of perishable foodstuffs and on the special equipment to be used for such carriage. United Nations Economic Commission for Europe. http://www.unece.org/fileadmin/DAM/trans/main/wp11/wp11fdoc/ATP-2011_final_e.pdf. Accessed: 15 September 2012.

Bøgh-Sørensen, L. (ed.) (2006) *Recommendations for the Processing and Handling of Frozen Foods*, 4th edn. International Institute of Refrigeration (IIR), Paris.

cheresources.com (2012) http://www.cheresources.com/content/articles/heat-transfer/u-in-heat-exchangers. Accessed: 15 September 2012.

Cleland, D.J., Cleland, A.C. and Earle, R.L. (1987) Prediction of freezing and thawing times for multidimensional shapes by simple formulae. Part 1: Regular shapes. *International Journal of Refrigeration*, **10**, 157–164.

Estrada-Flores, S. (2012) Transportation of frozen foods. In: *Handbook of Frozen Food Processing and Packaging*, 2nd edn (Sun, D.-W., ed.), Chapter 10. Marcel Dekker, pp. 217–233.

Fikiin, K., Tsvetkov, O., Laptev, Yu., Fikiin, A. and Kolodyaznaya, V. (2003) Thermophysical and Engineering Issues of the Immersion Freezing of Fruits in Ice Slurries Based on Sugar–Ethanol Aqueous Solution. Ecolibrium, August 2003, pp. 10–15.

German Insurance Association (GDV) (2012) *Container Handbook*. http://www.containerhandbuch.de/chb_e/wild/index.html. Accessed 15 September 2012.

Gill, C.O. (2012) Microbiology of frozen foods. In: *Handbook of Frozen Food Processing and Packaging*, 2nd edn (Sun,.Da-W., ed.), Chapter 7. Marcel Dekker, pp. 83–100.

Góral, D. and Kluza, F. (2012) Heat transfer coefficient in impingement fluidization freezing of vegetables and its prediction. *International Journal of Refrigeration*, **35**, 871–879.

Hossain, Md.M., Cleland, D.J. and Cleland, A.C. (1992) Prediction of freezing & thawing times for foods of regular multidimensional shape by using an analytically derived geometric factor. *International Journal of Refrigeration* **15**, 227–234.

Hung, Y.-C. and Kim, N.K. (1996) Fundamental aspects of freezing-cracking. *Food Technology*, **50**, 59–61.

International Institute of Refrigeration (IIR) (2008) Introduction Principles of RFID technologies for cold chain applications. 4th Informatory Note on *Refrigeration and Food*. International Institute of Refrigeration, Paris.

Kanda, Y., Aoki, M. and Kosugi T. (1992) Freezing of tofu (soybean curd) by pressure-shift: freezing and its structure. *Journal of Japan Society of Food Science and Technology*, **39**, 608–614.

Karel, M. and Lund, D.B. (2003) *Physical Principles of Food Preservation*, 2nd edn. Marcel Dekker.

Le Bail, A., Chevalier, D., Mussa, D.M. and Ghoul, M. (2002) High pressure freezing and thawing of foods: a review. *International Journal of Refrigeration*, **25**, 504–513.

Li, B. and Sun, D.-W. (2002) Effect of power ultrasound on freezing rate during immersion freezing. *Journal of Food Engineering*, **55**, 277–282.

Lin, Z., Cleland, A.C., Sellarach, G.F. and Cleland D.J. (1996a) A simple method for prediction of chilling times for objects of two-dimensional irregular shape. *International Journal of Refrigeration*, **19**, 95–106.

Lin, Z., Cleland, A.C., Sellarach, G.F. and Cleland, D.J. (1996b) A simple method for predictiion of chilling times: extension to 3-D irregular shapes. *International Journal of Refrigeration*, **19**, 107–114.

Meat and Livestock Australia (2012) *MLA Refrigeration Index Calculator*. http://www.foodsafetycentre.com.au/refrigerationindex.php. Accessed: 12 September 2012.

Miles, C.A., Mayer, Z., Morley, M.J. and Houska, M. (1997) Estimating the initial freezing point of foods from composition data. *International Journal of Food Science and Technology*, **32**, 389–400.

Orlowska, M., Havet, M. and Le Bail, A. (2009) Controlled ice nucleation under high voltage DC electrostatic field conditions. *Food Research International*, **42**, 879–884.

Pflug, I.J., Blaisdell, J.L. and Kopelman, I.J. (1965) Developing temperature–time curves for objects that can be approximated by a sphere, infinite plate or infinite cylinder. *ASHRAE Transactions*, **71**, 238–248.

Pham, Q.T. (1986) Freezing of foodstuffs with variations in environmental conditions. *International Journal of Refrigeration*, **9**, 290–295.

Pham, Q.T. (1987) Moisture transfer due to temperature changes or fluctuations. *Journal of Food Engineering*, **6**, 33–50.

Pham, Q.T. (2001) Prediction of cooling/freezing/thawing time and heat load. In: *Advances in Food Refrigeration* Sun, D.-W., ed.). Leatherhead Food RA, Leatherhead, pp.110–152.

Pham, Q.T. and Mawson R. (1997) Moisture migration and ice recrystallization in frozen foods. In: *Quality in Frozen Foods* (Erickson, M.C. and Y.C. Hung, Y.C. , eds). Chapman and Hall, New York, USA, pp. 67–91.

Pham, Q.T. and Oliver, D.W. (1983) Infiltration of air into cold stores. In: *Proceedings of the International Congress on Refrigeration*, Paris, vol. 4, pp. 67–72.

Pham, Q.T. and Willix J. (1984) A model for food desiccation in frozen storage. *Journal of Food Science*, **49**(1), 275–281 and 1294.

Plank, R. (1913) Beitrage zur Berechnung und Bewertung der Gefriergeschwindigkeit von Lebensmitteln. *Zeitschrift fur die gesamte Kalte Industrie*, **3**(10), 1–16.

Ramaswamy, H.S., Lo, K.V. and Tung, M.A. (1982) Simplified equations for transient temperatures in conductive foods with convective heat transfer at surface. *Journal of Food Science*, **47**, 2042–2047.

Reid, D.S., Kotte, K., Kilmartin, P. and Young, M. (2003) A new method for accelerated shelf-life prediction for frozen foods. *Journal of Science, Food and Agriculture*, **83**, 1018–1021.

Ruiz-Garcia, L., Loredana, L., Barreiro, P. and Robla, J.I. (2009) A review of wireless sensor technologies and applications in agriculture and food industry: state of the art and current trends. *Sensors*, **9**, 4728–4750.

Sinha, N.K. (2011) *Handbook of Vegetables and Vegetable Processing*. Blackwell, New York, USA.

Smale, N.J., Moureh, J. and Cortella, G. (2006) A review of numerical models of airflow in refrigerated food applications. *International Journal of Refrigeration*, **29**, 911–930.

Sun, D.-W (ed.) (2007) *Computational Fluid Dynamics in Food Processing*. CRC Press, Boca Raton, Florida, USA.

Sun, D.-W. and Wang, L.J. (2001) Vacuum cooling In: *Advances in Food Refrigeration*, Chapter 7. Leatherhead Publishers, Surrey, England, pp. 206–304.

Thompson, A.K. (2010) *Controlled Atmosphere Storage of Fruit and Vegetables*, 2nd edn. CAB International, Wallingford, England.

University of Nebraska–Lincoln (2012) UNL Models for Safety. http://numodels4safety.unl.edu/Pages/index.jsp. Accessed: 12 September 2012.

Visser, K. (1986) Automatic plate freezer development. *International Journal of Refrigeration*, **9**, 367.

16

Biotransformation in Food Processing

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16.1 Introduction

Biotransformation is the transformation of a substance A (substrate) to a substance B (product) using a biocatalyst (Kastner and Egerer, 1984). The essential difference between fermentation and biotransformation is that there are several catalytic steps between the substrate and the product in fermentation while there is only one or two in biotransformation. The distinction is also in the fact that the chemical structure of the substrate and the product resemble one another in a biotransformation, but not necessarily in fermentation (Liese, Steelbach and Wandrey, 2006; Kirk, Borchert and Fuglsang, 2002).

Biotransformation has the following advantages over chemical transformation:

- Reaction specificity
- Regiospecificity
- Stereospecificity
- Minimized side reactions
- Easier separation and fewer environmental problems
- Mild reaction conditions so harsh conditions like high temperature and pressure may be avoided, that is an energy and cost saving process.

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16.1.1 Different techniques of biotransformation

Biotransformations may be carried out using purified and immobilized enzymes, growing cultures, resting or washed microbial cells, immobilized cells and a combination of two sequential biotransformation steps catalysed by different microorganisms (Leuenberger, 1984).

Biotransformation with growing cells The substrate is added to the fermentation medium at the time of inoculation with a selected microbial strain or during a later phase of microbial growth if the substrate inhibits the cell growth. The optimum time of addition of the substrate must be determined; incubation is continued further until the maximum yield of transformation has been reached. The total time of biotransformation is relatively short because growth and biotransformation take place simultaneously.

Biotransformation with resting cells/washed cells The process is possible with endocellular enzymes. The microorganism is cultured under optimum growth conditions and harvested by centrifugation or filtration. The biomass is re-suspended in the substrate solution in water or buffer having the optimum pH for biotransformation. Supplementation of the transformation medium with easily metabolizable nutrients like glucose prolongs the viability and biotransformation activity of the cells and possible co-factor regeneration. The cell suspension is incubated at the optimized temperature until maximum product formation occurred. Use of the whole cell as an enzyme source has the following advantages:

(a) Laborious and costly process of enzyme purification may be avoided.
(b) As the enzymes remain within the cell, that is in their natural environment, they are more stable than in their isolated form (free form).
(c) If the biotransformation involved requires a co-enzyme, it is supplied and regenerated within the cell.

Biotransformation with an immobilized biocatalyst An immobilized biocatalyst (whole cell or enzyme) is preferred in the process of biotransformation as they have the following characteristics:

(a) Easy removal of the immobilized biocatalyst from the reaction mixture leads to repeated use.
(b) Continuous operation is possible by keeping the immobilized biocatalysts in the reaction vessel at a high biocatalyst concentration and maintaining a favourable substrate concentration for a long time.
(c) The product formation rate is high and inhibitory influences are minimal, providing maximum effective stability of the biocatalyst.

Biotransformation with purified enzymes or immobilized enzymes is preferred in the following cases:

(a) If the membrane of intact cells prevents proper substrate or product permeation.
(b) If degradation of products or undesirable side reactions takes place due to the presence of other enzyme systems.
(c) If the enzyme of interest is executed by the cell and can easily be purified from the medium after biomass removal.
(d) If the enzyme is available from animal and plant sources.
(e) If the enzyme of interest is available commercially.

16.2 Production of gluconic acid

Gluconic acid, a mild organic acid, is produced from glucose by an oxidation reaction (Figure 16.1). The reaction is facilitated by the enzyme glucose oxidase. Microbial production of gluconic acid has been the preferred method for several decades. The most studied and widely used fermentation process involves the fungus *Aspergillus niger*. Gluconic acids and its derivatives have wide applications in the food and pharmaceutical industries.

Gluconic acid is produced from glucose through a simple dehydrogenation reaction catalysed by glucose oxidase. Oxidation of an aldehyde group on the C-1 of β glucose to a carboxyl group results in the production of glucono-δ-lactone ($C_6H_{10}O_6$) and hydrogen peroxide. Glucono-δ-lactone is further hydrolysed to gluconic acid. The hydrolysis may be carried out spontaneously or by use of a lactone dehydrolysing enzyme, and hydrogen peroxide is decomposed to water and hydrogen. A purely chemical method may be used but the fermentation process is mostly preferred and has been used for several decades. The widely used fermentative method involves the production by *Aspergillus niger* (Ramachandran *et al*., 2006).

In 1980, Bourtoux reported that acetic acid bacteria are capable of producing sugar acid. Molliard in 1922 reported gluconic acid in *Sterigmatocystis nigra*, now known as *Aspergillus niger*. Production of gluconic acid by

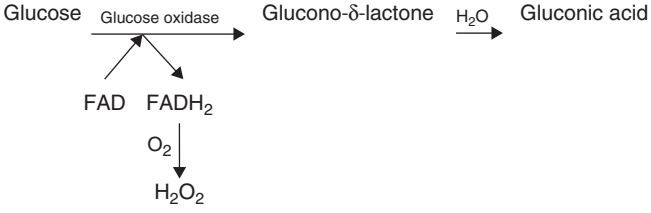

Figure 16.1 Production of gluconic acid using glucose oxidase

bacterial species viz. *Pseudomonas*, *Gluconobacter*, *Acetobacter* and various fungal strains was also reported (Molliard, 1922).

Current commercial production of sodium gluconate uses submerged fermentation with *A. niger* and is based on the modified process developed by Blom *et al.* (1952). It involves fed-batch cultivation with intermittent glucose feedings and the use of sodium hydroxide as the neutralizing agent. The pH is held at 6.0–6.5 and the temperature is held at about 34 °C. The productivity of this process is very high since glucose is converted at a rate of 15 g/L h.

Gluconic acid is used in various fields such as food, chemical analysis, mainly as a glucose sensor, and in the quantitative determination of glucose in body fluids and urine. It is used in food processing in the removal of glucose prior to the preparation of products such as dried eggs to reduce nonenzymatic browning. It is also used in removing residual oxygen from fruit juices, beer and wine, and also from dehydrated packaged foods.

16.2.1 Gluconic acid production by filamentous fungi

Glucose oxidase is a flavour protein that contains one very tightly but noncovalently bound flavin adenine dinucleotide (FAD) cofactor per monomer and is a homodimer with a molecular mass of 130–320 kDa depending on the extent of glycosylation. It catalyses the reaction where glucose is dehydrated to glucono-δ-lactone, while hydrogen is transferred to FAD by transmission of the hydrogen to oxygen to form hydrogen peroxide. Glucose oxidase is a glycoprotein. The native enzyme is glycosylated with a carbohydrate mass percentage of 16–25% (Swoboda and Massey, 1965; Pazur and Kleppe, 1964).

The enzyme is induced in the presence of high levels of glucose in the medium, pH around 5.5 and an elevated oxygen level. The enzyme is stable between pH 4.0 and 6.0 at 40 °C for 2 h but is unstable above 50 °C. Liu *et al.* (2001) conducted a study on the effect of metal ions on the simultaneous production of glucose oxidase and catalase and found that calcium carbonate induced the synthesis of both enzymes. The induction of calcium carbonate was accompanied by a metabolic shift from the glycolytic pathway to direct oxidation of glucose by the enzyme. The enzyme is found to be inhibited by hydrogen peroxide, the by-product of gluconic acid production.

16.2.2 Production by *Aspergillus niger*

Aspergillus niger produces all the enzymes required for the conversion of glucose to gluconic acid, which include glucose oxidase, catalase, lactonase and mutarotase, although crystalline glucose monohydrate, which is in the alpha form, is converted spontaneously into the beta form in the solution. *A. niger* also produces the enzyme mutarotase, which serves to accelerate the reaction (Cho and Bailey, 1977).

Production of gluconic acid is directly linked with glucose oxidase activity. Depending on the application, the fermentation broths containing sodium gluconate or calcium gluconate are produced by the addition of solutions of sodium hydroxide or calcium carbonate, respectively, for neutralization. The general optimal conditions for gluconic acid production are as follows (Rohr, Kubicek and Komineck, 1983): glucose concentration is 110–250 g/L, nitrogen phosphorus sources are used at a very low concentration (20 mM), the pH value of the medium is between 4.5 and 6.5 and a very high aeration rate is achieved by application of an elevated air pressure (4 bar).

Oxygen availability and pH of the culture medium are the two key parameters that influence gluconic acid production. Glucose oxidase uses molecular oxygen in the bioconversion of glucose. Gluconic acid production is an extremely oxygen consuming process with a high oxygen demand for the bioconversion reaction, which is strongly influenced by the dissolved oxygen concentration. Oxygen is generally supplied in the form of atmospheric air; however, in some studies, high pressure pure oxygen has also been provided. Sakurai *et al*. (1989) reported that high pressure oxygen at about 6 bar was supplied and maintained the dissolved oxygen level at 150 ppm; pH is another important parameter for the production of gluconic acid. The optimum pH for production of gluconic acid by *A. niger* is around 4.5 to 7.0 (Znad, Markos and Bales, 2004; Lockwood, 1979).

16.2.3 Production and recovery of gluconic acid by immobilized biomass

The bioconversion of glucose to gluconic acid was studied by Sankpal *et al*. (1999) using *A. niger* immobilized on cellulosic fabric. Glucose solution (100 g/L) was passed through capillaries of a vertical fabric support and oxidized to gluconic acid (120–140 g/L) at the interface. The concentration was higher than the expected value of 109 g/100 g of glucose due to evaporative concentration during the downflow. The system was continuously run for 61 days (Sankpal and Kulkarni, 2002; Sakurai *et al*., 1989).

For obtaining calcium gluconate as a product, calcium hydroxide or calcium carbonate is used as the neutralizing agent. They are added to the nutritive broth accompanied by heating and vigorous stirring. The broth is concentrated to a hot supersaturated solution of calcium gluconate, followed by cooling at 20 °C and adding water-miscible solvents, which crystallizes the compound. A treatment with activated carbon facilitates the crystallization process. Finally, they are centrifuged, washed several times and dried at 80 °C.

For the recovery of free gluconic acid from calcium gluconate the broth is clarified, decolourized, concentrated and exposed to −10 °C in the presence or absence of alcohol. Thus, the calcium salt of gluconic acid crystallizes and is then recovered and further purified. Gluconic acid can also be obtained by

precipitating the calcium gluconate from hypersaturated solution in the cold and released subsequently by adding sulfuric acid stoichiometrically, removing the calcium as calcium sulfate. Another method of passing the solution through a column containing a strong cation is also practised where the calcium ions are absorbed.

Sodium gluconate, the principal manufactured form of gluconic acid, is prepared by ion exchange. In the process developed by Blom *et al*. (1952), the sodium gluconate from the filtered fermented broth is concentrated to 45% (mass/volume), followed by the addition of sodium hydroxide by rising the pH to 7.5 followed by drum drying. Carbon treatment of the hot solution before the drying process is practised for obtaining a refined product. Glucono-δ-lactone recovery is a simple process. An aqueous solution of gluconic acid is an equilibrium mixture of glucono-δ-lactone, glucono-lactone and gluconic acid. At a temperature between 30–70 °C, the crystal that is separated from the supersaturated solution is glucono-δ-lactone. At temperatures below 30 °C, gluconic acid results even above 70 °C and the resulting product would be glucono-lactone.

16.3 Ascorbic acid

L-Ascorbic acid (L-AA) or vitamin C is a water-soluble vitamin that is essential for some physiological functions. It was first isolated from adrenal glands (Szent-Gyorgyi, 1928) and subsequently characterized from plant tissues (Herbert *et al*., 1933; Svirbely and Szent-Gyorgyi, 1932; Waugh and King, 1932). L-AA is an essential nutrient for humans, nonhuman primates and a few other mammals (Simronoff, 2001; Sauberlich, 1994) as they cannot synthesize this vitamin.

Due to its antioxidant properties and its potential to stimulate collagen production (Lupo, 2001), L-AA is required in the pharmaceutical industry. It is used as a component of vitamin supplements, an additive to cosmetic products and in pharmaceutical preparations for the treatment of burns. It has an unexpected role in cancer treatment. However, intravenous injections are necessary as the doses are so high (Chen *et al*., 2005). Beverages and food industries exploit the general antioxidant properties of L-AA. In the food industry, L-AA is also employed to prevent pigment discolouration and enzymatic browning (Mihalev *et al*., 2004; Pilizota and Subaric, 1998), to protect flavour (Bauernfeind and Pinkert, 1970), to enhance nutrient contents (Hancock and Viola, 2005; Lindley, 1998) and to extend shelf-life (Chauhan, Ramteke and Eipeson, 1998).

Originally, L-AA was isolated from lemons. The first chemical L-AA synthesis process from L-xylosone was achieved in 1933 (Haworth and Hirst, 1933; Reichstein, Grussner and Oppenauer, 1933). A biochemical step was introduced that allowed the preparation from D-glucose (Reichstein and Grussner,

1934). At present, a considerable part of commercially manufactured L-AA is still synthesized via the seven-step Reichstein process, which begins with the chemical hydrogenation of D-glucose to form D-sorbitol. The initial step is followed by a biological reaction catalysed by *Gluconobacter oxydans*, which regiospecifically oxidized D-sorbitol to L-sorbose.

Since only the isomer is biologically active, the stereospecific biotransformation using *G. oxydans* is highly important as protection group chemistry could be avoided. The product L-sorbose is then crystallized and after condensation with acetone, sorbose-diacetone is formed, which is afterwards oxidized to 2-keto-L-gluconic acid (2-KLGA) using a platinum catalyst. Subsequent to enolization and lactonization, L-AA is obtained with a final yield of about 50% (Boudrant, 1990). The Reischstein process has gained all the advantages that would be expected after such a long period of application and development. The process is still an energy consuming process as high temperatures and pressures are necessary for some steps of the process. Hence, there exists a tremendous interest to search for an alternative process. As a result, microbiological biotransformations using reasonable raw materials became the focus of particular attention.

The genetically engineered strains accumulate more or less high amounts of 2-KLGA. However, none of these strains directly converts D-glucose, D-sorbitol or their oxidation products to L-AA. The enzymes capable of the conversion of 2-KLGA to L-AA are of major interest. Recently, Berry *et al.* (2005) reported that L-sorbosone dehydrogenase is responsible for the conversion of L-sorbosone to L-AA. The corresponding gene was over-expressed in the same strain. In resting cell experiments using the recombinant strain, 4.2 g/L of L-AA were produced from 10 g/L of L-sorbosone; the identity of the L-AA was confirmed by high-performance liquid chromatography (HPLC)–mass spectrometry (MS) (Berry *et al.*, 2005).

As presented above, so far the bioconversion of D-glucose or D-sorbitol to L-AA in terms of an industrial process is still under way. Therefore, further improvement of the available conversion process is necessary for optimization of gene expression, fermentation process, elimination of by-product formation and carbon flux redirection; it is also necessary to improve the conversion process.

16.4 Lactose hydrolysis by β-galactosidase

β-Galactosidase hydrolyses milk sugar lactose into glucose and galactose (Figure 16.2) and also catalyses the synthesis of different galactosides. β-Galactosidase finds wide application in the three main areas of food technology, health and environment. Lactose is the main sugar found in milk and whey (~4.2%). Whey is the liquid that remains after the solid component has been removed from the cream or milk during cheese production.

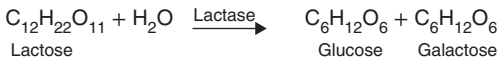

Figure 16.2 Lactose hydrolysis by β-galactosidase

Many individuals all over the world (70–90% Asian and Africans and 2–15% Americans and West Europeans) have β-galactosidase deficiency and suffer from lactose intolerance syndrome after consumption of milk due to insufficient amounts of β-galactosidase in the small intestine to hydrolyse the disaccharide lactose. As a result, unhydrolysed lactose arrives in the large intestine, resulting in several disorders such as flatulence, abdominal bloating, cramps and diarrhea. The β-galactosidase enzyme may be used to reduce the lactose content of milk.

The high lactose content in milk leads to the production of inferior quality ice cream and condensed milk due to crystallization of lactose. This problem can be easily overcome by hydrolysing lactose using the enzyme prior to condensation of milk. Moreover, glucose and galactose are much sweeter than lactose. The use of β-galactosidase for hydrolysis of lactose may result in improved dairy products and processes. Pre-hydrolysis of milk with lactase would shorten yogurt and cheese production time by 20% as well as increase the yield and sweetness. A significant reduction in manufacturing time, ripening time and improved quality has already been observed for different cheeses. For all these reasons, the applications of β-galactosidase are increasing steadily in the sweet meat industry. Lactose present in whey (BOD ie biochemical oxygen demand value is 30 000–60 000 mg/L) is mainly responsible for pollution. Glucose and galactose may be produced from lactose using β-galactosidase for subsequent production of different useful substances while simultaneously reducing the BOD value of whey.

For the production of low lactose or lactose-free milk, the use of free β-galactosidase is not cost effective, as after lactose hydrolysis the enzyme becomes inactivated by heat treatment. To overcome this problem, immobilized enzyme may be used for the hydrolysis instead of free enzyme (Panesar, Kumari and Panesar, 2010). Central del Latte of Milan (Italy) was the first to develop commercial hydrolysis of lactose in milk by using immobilized *Saccharomyces (Kluyveromyces) lactis* lactase entrapped in cellulose triacetate fibre. Drouin Co-operative Butter Factory used β-galactosidase of fungal origin immobilized on a rugged surface of an amphoteric ion exchange resin of phenol formaldehyde polymer (developed by Sumitomo Chemical, Japan) to produce market milk and hydrolysed whey (Panesar, Kumai and Panesar, 2010; Ray and Gupta, 2003). Snow Brand Factory developed a rotary column reactor that could be used both as a stirred tank reactor and as a packed bed reactor (Honda *et al*., 1991). The reaction rate was greatly affected by packing density of the immobilized enzyme. This reactor can also overcome

the problem of channelling or a severe pressure drop. A 70–80% lactose hydrolysis was observed in horizontal rotary column and washing of immobilized enzyme was carried out for 36 cycles, indicating that the process is well suited for hydrolysis of lactose in milk (Panesar, Kumari and Panesar, 2010).

Thus, immobilized β-galactosidase technology is an effective process for successful hydrolysis of lactose and it is also cost effective. The major problems associated with the immobilized enzyme system are microbial contamination, protein adherence and channelling. Hence, for long term operations with immobilized system, periodic washing and pasteurization are indispensible processes (Hirohara *et al*., 1982).

16.5 Invert sugar

Inverted sugars are sweeter compared to sucrose and tend to retain moisture and are less prone to crystallization. Inverted sugars are therefore valued by bakers. Sucrose may be hydrolysed to glucose and fructose by simple heating but catalysts are added to accelerate the conversion reaction. Sucrases (in animals) and invertases (in plants) are the biological catalysts (glycoside hydrolase enzymes) used for this purpose. Lemon juice or cream of tartar are used as acid to accelerate the inversion of sucrose. Inverted sugar syrup can be easily made by adding roughly one gram of citric acid or ascorbic acid per kilogram of sugar. Cream of tartar (one gram per kilogram) or fresh lemon juice (10 mg per kilogram) may also be used.

The term 'inverted' is derived from the method of measuring the concentration of sugar syrup using a polarimeter. When polarized light is passing through a sample of pure sucrose solution, it is rotated to the right (optical rotation). After inversion of sucrose, the direction of rotation changes (inverted) from right to left. $C_{12}H_{22}O_{11}$ (sucrose, specific rotation = +66.5°) + H_2O (water, no rotation) → $C_6H_{12}O_6$ (glucose, specific rotation = +52.7°) + $C_6H_{12}O_6$ (fructose, specific rotation = −92°); net: +66.5° converts to −39.3°.

The mixture is boiled for 20 min to get a temperature of 114 °C and it converts enough of the sucrose to effectively prevent crystallization, without giving a noticeably sour taste. Invert sugar syrup may also be produced without the use of acids or enzymes by thermal means alone; two parts of granulated sucrose and one part of water simmered for five to seven minutes will convert a modest portion to invert sugar.

All inverted sugar syrups are created from hydrolysing sucrose to glucose (dextrose) and fructose by heating a sucrose solution and then relying on time alone, with the catalytic properties of an acid or enzymes used to speed the reaction. Commercially prepared acid catalysed solutions are neutralized when the desired level of inversion is reached. All constituent sugars (sucrose, glucose and fructose) support fermentation and thus invert sugar solutions may be fermented as readily as sucrose solutions.

Invert sugar has a lower water activity than that of sucrose and provides more powerful preserving qualities (a longer shelf-life) to products that use it. The shelf-life of partial inverts is approximately six months, depending on the storage and climatic conditions. Crystallized invert sugar solutions may be resorted to their liquid state by gentle heating.

16.5.1 Use of enzyme or cell

Fungal biomass of *Cladosporium cladosporioides* may be used as a natural source of invertase. It is a low-cost technology. This offers great operational stability to the enzyme, allowing it to be reused without significant loss in activity (Coutinho Filho, Hori and Ribiero, 1999; Costaglioli *et al*., 1997). Invertase or β-D-fructofuranosidase (E.C.3.2.1.26) has been used for a long time for both bulk conversion of sucrose and in situ hydrolysis of sucrose in confectionery. Usually the enzyme is obtained from baker's yeast (*Saccharomyces cerevisiae*). The isolated enzyme can be used in the free as well as the immobilized form (Tanriseven and Dogan, 2001). Isolation and purification of the enzyme increases the cost of invertase.

16.5.2 Use of immobilized cells

Soluble invertase from *S. cerevisiae* as well as the whole biomass may be used to obtain invert sugar (Linko, Weckstro and Linko, 1980). A number of researchers studied the immobilization of invertase (MW = 27000 Da) using alginate. Ro and Kim (1991) reported that 80% of the invertase is leaked out when immobilized in alginate beads. Husain, Iqbal and Saleemuddin (1985) reported that coupling of invertase to concavalin-A before entrapment into alginate may prevent the leakage. Aruda and Vitolo (1999) studied the effect of pH on immobilization of invertase and found that the beads retained 50 and 60% of the enzyme if they were formed at pH 4.0 and 8.0, respectively.

The use of the whole cell of *S. cerevisiae* as a biocatalyst may lower the production cost. Biomass may be immobilized using alginate, gelatin (Parascandola and Scardi, 1982) and modified jute fabric. The particle size of the biocatalyst significantly influences the catalytic activity. The diameter of many biocatalysts usually ranges between several hundred micrometres and a few millimetres (Rebros *et al*., 2007), which enables simple separation of the biocatalyst from the reaction mixture. Smaller particles of immobilized yeast cells can lead to an increase in the conversion rate. Magnetically responsive cell-containing beads are of special interest as they may work in difficult-to-handle samples, such as suspensions (Safarikova and Safarik, 2001; Safarik and Safarikova, 1997). Magnetic properties of the beads are usually caused by the presence of magnetic iron oxide nano- and microparticles.

16.6 Production of oligosaccharides

The enzyme β-galactosidase can also be used in transglycosylation of lactose to synthesize galacto-oligosaccharides (GOSs). These are nondigestible oligosaccharides and are not hydrolysed or absorbed in the upper intestinal tract; they pass on to the colon where they are fermented selectively by beneficial intestinal bacteria. Oligosaccharides and *Bifidobacteria* provide a wide variety of health benefits, including anticarcinogenic effects, reduction in serum cholesterol, improved liner function, reduction of colon cancer risk and improved intestinal health (Hawkins, 1993).

16.6.1 Synthesis of galacto-oligosaccharides

β-Galactosidase also has transferase activity and can produce and hydrolyse a series of oligosaccharides, which have a beneficial effect on the growth of desirable intestinal microflora (Mahoney, 1998). The transferase reaction can be used to attach galactose to other chemicals, resulting in the formation of galacto-oligosaccharides (GOSs), and consequently has potential application in the production of food ingredients, pharmaceuticals and other biologically active compounds. The reaction conditions for transgalactosylation are the high lactose concentration, elevated temperature and low water activity in the reaction medium (Zarate and Lopez-Leiva, 1990).

Galacto-oligosaccharides (GOSs) from lactose were produced by partially purified β-galactosidase from *Bullera singularis* ATCC 24193 immobilized on Chitopearl BCW3510 bead in a packed bed reactor, which resulted in 55% (w/w) oligosaccharides with a 15 day operation (Albayrak and Yang, 2002).

16.7 Glucose isomerization

Isomerization of glucose may be carried out chemically under mild alkaline conditions. During isomerization, other sugar ketones are produced as by-products. Marshall and Kool (1957) reported that the enzyme glucose isomerase is capable of coverting glucose to fructose. The producing organisms are *Actinoplanes missouriensis*, *Bacillus coagulans*, *Streptomyces olivaceus* and *S. olivochromogenes*.

The optimum pH and temperature for glucose isomerases produced by different strains are more or less similar. The optimum conditions for immobilized glucose isomerase from *B. coagulans* are a pH range of 7–8, temperature of about 80 °C and reaction time of 15 min. For use in industries a temperature of 60 °C is chosen as a useful compromise between stability and enzyme activity. Mg ion stabilizes the enzyme. The requirement for the Mg ion depends on the Ca content of the 96 DE corn syrup. About 0.0004 M of $MgSO_4$ is required if the Ca content is less than 1 ppm (Peppler and Reed, 1984).

The continuous isomerization of glucose (96 dextrose equivalent corn syrup) to fructose using immobilized glucose isomerase in a packed bed column is generally carried out under the following conditions: inlet pH of 8.2, temperature of 60 °C and hold-up time of 0.5 to 4.0 h depending on the size of the bed and enzyme activity. The conversion is carried out to a fructose concentration of only 42–43% (based on solid) due to the slow rate of attaining equilibrium. The concentration of the high DE syrup fed into the packed bed is usually 40–45% solid. Immobilized glucose isomerase enzyme columns are used continuously for several months. The isomerization reaction reaches equilibrium at a fructose concentration of about 55%. Higher fructose levels can be achieved by fractionation of the 42% fructose syrup in chromatographic columns. Fructose is more strongly adsorbed by the cationic group of the column. The first effluent containing glucose and only about 1.5% fructose is recirculated to the isomerization unit. Then fructose (85% concentration) is recovered from the chromatographic column. This is mixed with HFCS 42 to give HFCS 55. The production process scheme is shown in Figure 16.3.

Three high fructose corn syrup (HFCS) products are commercially available. These products contain 42, 55 and 90% fructose. In most applications, HFCS replaces sucrose as the sweetener. The 42% fructose can be used as a liquid sweetener in most food products (beverages, processed foods, cereals and baked goods). The most widely used variety, HFCS with 55% fructose, is used in soft drinks. HFCS with 90% fructose is an ideal sweetener for reduced calorie foods such as jams and jellies and is primarily used to blend HFCS 42 to make HFCS 55. The relative sweetness of HFCS 55 is comparable to table sugar (sucrose), a disaccharide of fructose and glucose. HFCS 42 is less sweet than sucrose while, being a liquid, it is easier to blend and transport.

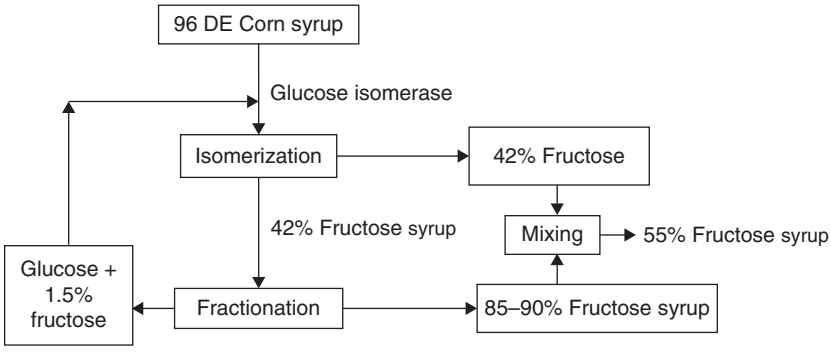

Figure 16.3 Production process of high fructose syrup

16.8 Production of flavour and fragrance

Food processing operations, from premature harvesting to extended storage and physical treatments, may cause a loss of flavour that calls for subsequent supplementation. In addition, the steadily increasing market for flavours forces suppliers to search for alternative sources. Biotechnological generation of flavour compounds is becoming increasingly attractive (Krings and Berger, 1998).

16.8.1 Monoterpenes

Monoterpenes are widely distributed in nature (more than 400 structures) and constitute suitable precursor substrates. Soil bacteria and filamentous fungi transform acyclic, monocyclic and bicyclic monoterpenoids. Reviews of isoprenoid biosynthesis, *de novo* generation and opportunities for microbial biotransformation have been published recently (Breheret *et al*., 1997; McCaskill and Croteau, 1996; Van der Werf, De Bont and Leak, 1996; Seitz, 1994).

Most of the monoterpene biotransformation studies described so far have been of more academic than practical value, and no monoterpene biotransformation process has been commercialized yet. Major problems encountered are that both precursor (monoterpene) and product (terpenoid) are chemically unstable, monoterpene precursors have low water solubility, precursors and products are both highly volatile, both precursors and products have high cytotoxicity and the transformation rate is low.

16.8.2 Curcumin to vanillin

Vanillin (4-hydroxy-3-methoxybenzaldehyde) is quantitvely one of the most important aromatic flavour additives in the world and is used widely in foods, beverages, perfumes, pharmaceuticals and medicinal industries (Serra, Fuganti and Brenna, 2005; Keshava *et al*., 1998).

The production of vanillin by bioconversion has been extensively studied in the past and biocatalysts employed have included fungi, bacteria, genetically engineered microorganisms, plant cells and enzymes extracted from biological materials (Mabinya, Mafunga and Brand, 2006) and several potential feed stocks have also been suggested including phenolic stilbenes, eugenol (Chatterjee, De and Bhattacharyya, 1999) and ferulic acid (Ghosh, Sachan and Mitra, 2006; Converti *et al*., 2003; Lin and Lin-Shiau 2001). *Rhodococcus* strains have been reported to produce vanillin from ferulic acid and eugenol (Serra, Fuganti and Brenns, 2005). It is also produced

from curcumin by *Rhodococcus rhodochrous* MTCC 265 (Bharti and Gupta, 2011). These authors used the seed culture medium having composition: peptone 0.5%, sodium chloride 0.5%, beef extract 0.1% and yeast extract 0.2%, and the flask was incubated at 30 °C and 180 rpm. A 10% aliquot of seed culture was used to inoculate 50 mL of minimal salt medium (MSM) at pH 7 containing 0.07% K_2HPO_4, 0.03% KH_2PO_4, 0.05% $MgSO_4.7H_2O$, 0.025% $(NH_4)_2SO_4$, 0.001% $FeSO_4.7H_2O$, 0.0001% $ZnSO_4.7H_2O$, 0.0001% $MnSO_4$ and different concentrations of sterilized glucose (0.2–0.5%), yeast extract (0.01–0.1%) and peptone (0.03–0.1%); 3.56 mg/L of vanillin was produced from 29.02 mg/L of curcumin in this experiment.

16.8.3 Benzaldehyde production

Benzaldehyde is the second most important flavour after vanillin. Natural benzaldehyde is usually liberated from amygladin, a cyanogenic glycoside present in fruit kernels, and is used as a key ingredient in cherry and other natural fruit flavours. The microbial degradation of natural phenylalanine offers an alternative. This process is aided by a plentiful cheap supply of natural L-phenylalanine, which has become available as an intermediate of the synthesis of the high intensity sweetner, aspartem (Cheethan, 1996).

The whole cells of the microorganism were used as a source of intracellular enzyme, avoiding cell disintegration and complex purification of the enzyme. Benzoylformate was converted to benzaldehyde using both the free whole-cell enzyme (benzoylformate decarboxylase) from *Pseudomonas putida* cells and the whole-cell enzyme immobilized in a calcium alginate liquid-core capsule. Cells inoculated in calcium alginate capsules were cultured for 2 days in a medium containing a large amount of nitrogen sources and then cultured successively for 1 day in a new medium containing a large amount of phosphate. In a free whole-cell enzyme survey, benzaldehyde produced by the catalysis of whole-cell benzoylformate decarboxylase was consecutively converted to benzyl alcohol by benzyl alcohol dehydrogenase. Encapsulation of whole-cell enzymes gave a resistance of mass transfer in a capsule membrane that caused a lagging of appearance of benzaldehyde in the reaction medium. Encapsulated whole-cell enzyme from *P. putida* could be re-used to produce benzaldehyde from benzoylformate without formation of benzyl alcohol and retained 60% of the initial activity after 30 batches.

Benzaldehyde formation in a medium supplemented with phenylalanine has been reported for cultures of *Ischnoderma benzoinum* (Fabre, Blanc and Goma, 1996; Krings, Hinz and Berger, 1996), *Polyporus tuberaster* (Kawabe and Morita, 1994) and *Phanerochaete chrysosporium* (Jensen *et al*., 1994). Immobilization of the white-rot fungus *Bjerkandera adusta* resulted in an increased production of benzaldehyde in a medium containing l-phenylalanine (Lapadatescu *et al*., 1997). Among bacteria, benzaldehyde formation has been reported for a strain of *Pseudomonas putida*. In this strain, benzaldehyde was formed as a metabolic intermediate in the mandelic

acid pathway during the degradation of mandelic acid (Tsou, Ransom and Gerlt, 1990). Several metabolic pathways have been proposed in the literature for the formation of benzaldehyde from phenylalanine.

16.9 Artificial sweetener

16.9.1 Xylitol

In nature, D-xylitol is found in many fruits and vegetables viz. berries, corn husks, oats, lettuces, cauliflowers, mushroom, etc. (Chen *et al*., 2010). It is non-carcinogenic and may be used as food additives and sweetening agent (sucrose substitutes) to replace sucrose especially for noninsulin diabetics. It prevents dental caries and is completely safe for teeth. Xylitol is approved for usages in foods, pharmaceuticals and oral health products in more than 35 countries (Rao, Jothi and Rao, 2008).

Microbial conversion of D-xylose to D-xylitol In bacteria, the conversion of D-xylose to D-xylulose is catalysed by xylose isomerase in a single step. The xylose isomerase was also detected in some yeasts and moulds such as *C. boidinni*, *Malbranchea pulchella* and *Meurospora crassa*. However, in the majority of yeast and fungi, the conversion of D-xylose to D-xylulose needs two steps, a reduction step followed by an oxidation step. D-Xylose was first reduced to D-xylitol by either NADH or NADPH-dependent xylose reductase aldose reductase (E.C.1.1.1.21) (XR); the resulting D-xylitol was either secreted or further oxidized to D-xylulose by NAD- or NADP-dependent xylitol dehydrogenase (EC.1.1.1.9) (XDH). These two reactions are considered to be the rate-limiting steps in D-xylose fermentation and D-xylitol production. Some strains of yeast could metabolize D-xylulose to D-xylulose-5-phosphate by xylulokinase (E.C. 2.7.1.17) (XK). Xylulose-5-phosphate can subsequently enter the pentose phosphate pathway (Lachke and Jeffries, 1986; Smiley and Bolen, 1982).

16.9.2 Mannitol

Mannitol is about 50% as sweet as sugar. It is manufactured for use in food and pharmaceuticals and is found naturally in mushrooms and trees. Research efforts have been directed towards the production of mannitol by fermentation and enzymatic process (Ghoreishi and Shahrestani, 2009; Song and Vielle, 2009; Saha and Racine, 2008; Vandamme and Soetaert, 1995).

Lactic acid bacteria (LAB), yeast and fungi are known to produce mannitol from fructose or glucose (Saha, 2003; Song *et al*., 2002; Wisselink *et al*., 2002; Smiley, Cadmus and Liepins, 1967). Both homo- and heterofermentative LAB produce mannitol (Saha, 2003). Some homofermentative LAB such as *Streptococcus mutants* and *Lactobacillus leichmanii* produce small amounts of mannitol from glucose (Chalfan, Levy and Mateles, 1975).

A number of heterofermentative LAB of the genera *Lactobacillus*, *Leuconostoc* and *Oenococcus* can produce mannitol directly from fructose (Saha, 2003). *L. fermentum* NRRL B-1915, *L. intermedius* NRRL B-3693, *L. Amelilibiosum* NRRL B-742, *L. Citrovorum* NRRL B-1147, Leu. Mesenteroides subsp. dextranicum NRRL B-1120 and *L. Paramesenteroides* NRRL B-3471 produce mannitol from fructose. The strain *L. intermedius* NRRL B-3693 produced 198 g of mannitol from 300 g of fructose/L in pH-controlled (pH 5.0) fermentation at 37 °C. The time of maximum mannitol production varied greatly from 15 h at 150 g fructose to 136 h at 300 g fructose/L. The bacterium converted fructose to mannitol during the early growth stage. A competitive production process for mannitol by fermentation would require inexpensive raw materials. Saha (2006) studied the production of mannitol by *L. intermedius* NRRL B-3693 using molasses as a carbon source. The bacterium produced mannitol (104 g/L) from a mixture of molasses and fructose syrup (1:1) (total sugars, 150 g/L; fructose/glucose, 4:1) in 16 h. Several kinds of inexpensive organic and inorganic nitrogen sources and corn steep liquor (CSL) were evaluated. Several filamentous fungi produced mannitol from glucose.

The fungal strain converted glucose to mannitol with a 50% yield based on glucose consumed in 10–16 days by feeding glucose daily with a volumetric productivity of 0.15 g/L h and a yield of 31.0 mol%. The presence of glucose in the medium was essential to prevent metabolism of mannitol. Lee (1967) determined the carbon balance for fermentation of glucose to mannitol by *Aspergillus* sp. The products found were: cells (17% of carbon input), CO_2 (26%), mannitol (35%), glycerol (10%), erythritol (2.5%), glycogen (1%) and unidentified compounds (8%).

16.9.3 Sorbitol

Sorbitol, a polyol ($C_6H_{14}O_6$), also known as D-glucitol, having 60% sweetness compared to sucrose, is found in many fruits such as berries except grapes, cherries, pulms, pears and apples (Budavari *et al*., 1996; Wrolstad and Shallenberger, 1981). At present, it is produced chemically. The bacterium *Zymomonas mobilis* is able to produce sorbitol and gluconic acid from fructose and glucose, respectively.

It is used in confectionery, chewing gum, candy, dessert, ice cream, diabetic food and a wide range of food products, not only as a sweetener but also as a humectant, texturizer and softener. Other applications include pharmaceutical products, sorbose, ascorbic acid, propylene glycol, synthetic plasticizers and resins, etc. (Budavari *et al*., 1996; Elvers, Hawkins and Russey, 1994; Albert, Stratz and Vollheim, 1980). More than 50% of the sorbitol produced is used as 70% sorbitol solution and about one-quarter is used for the synthesis of vitamin C.

Biotechnological production of sorbitol Considering the low yield of sorbitol obtained in the conventional fermentation method, Chun and Rogers (1988) attempted to use previously grown (washed cells), concentrated and permeabilized (with 10%, v/v) cells of *Z. mobilis* ZM4 (ATCC 31821) for sorbitol production. The essential soluble co-factors are released during permeabilization. In a 16 h batch process using free tolune-treated cells, the yield of sorbitol and gluconic acid were 290 and 283 g/L, respectively. Yields were about 95% for both products. Similar results were obtained using tolune-treated cells immobilized in Ca-alginate beads. Some enzyme activity is lost if the beads are re-utilized. A 125 h continuous operation was also reported using immobilized cells having 80–85 g/L with productivities of 7.6 and 7.2 g/L h, respectively (Scopes, Rogers and Leigh, 1988).

Bringer-Meyer and Sahm (1991) reported and patented an alternative method for cell (*Z. mobilis*) permeabilization by freezing at −20 °C and thawing at room temperature. The yield was close to 100% for both sorbitol and gluconic acid.

The problems regarding the industrial bioproduction of sorbitol included the relatively high cost of substrate, particularly fructose, compared to the product value. Ro and Kim (1991) studied the bioconversion of sucrose to sorbitol and gluconic acid using toluene-treated *Z. mobilis* and invertase co-immobilized in both chitin and calcium alginate. The optimum substrate concentration for the co-immobilized enzymes in both chitin and calcium alginate was determined as 200 g/L.

Most of the reported results involved permeabilized *Z. mobilis* cells. However, the use of nonpermeabilized cells of *Z. mobilis* was also proposed (Silveira *et al*., 1999; Silveira, Lopes da Costa and Jonas, 1994). Attempts have been made to develop a functional process for the biotechnological production of sorbitol and gluconic acid (Silva-Martinez *et al*., 1998; Nidetzky *et al*., 1997; Gollhofer *et al*., 1995). Silveira, Lopes da Costa and Jonas (1994) proposed a method for recovering sorbitol and sodium gluconate by selective precipitation with organic solvents like methanol and ethanol. Recently, Ferraz, Alves and Borges (2001) reported the use of an electrodialysis system coupled to the bioreactor to simultaneously remove gluconic acid from the medium as it is produced. Although good results were described for these methods on the laboratory scale, a cheap and efficient method has to be developed and optimized for an industrial process.

16.10 Conclusions

Biotransformation using microbial biomass and enzymes is becoming an important tool in food processing technology. Biotransformations are most useful when a given reaction cannot be easily performed by chemical methods. The process has a number of advantages over chemical processes,

viz. reaction specificity, stereospecificity, mild reaction conditions, minimum side reaction, easy product recovery and an ecofriendly system. Continuous production is also possible at industrial levels using an immobilized biomass or enzyme. Several food products are at present developed commercially using biotansformation processes while many other biotransformation processes are still in the exploratory phase.

References

Albayrak, N. and Yang, S.T. (2002) Production of galactosides from lactose by *Aspergillus oryzae β*-galactosidase immobilized on cotton cloth. *Biotechnology and Bioengineering*, **77**(1), 8–9.

Albert, R., Stratz, A. and Vollheim, G. (1980) Die katalytische Herstel-lung von Zucker-alkokolen und deren Verwendung. *Chemie Ingenieur Technik*, **52**, 582–587.

Arruda, L.M.O. and Vitole, M. (1999) Characterization of invertase entrapped into calcium alginate beads. *Applied Biochemistry Biotechnology*, **81**(1), 23–33.

Bauernfeind, J.C. and Pinkert, D.M. (1970) Food processing with added ascorbic acid. *Advances in Food Research*, **18**, 219–315.

Berry, A., Lee, C., Mayer, A.F. and Shinjoh, M. (2005) Microbial production of l-ascorbic acid. World Intellectual Property Organisation, Patent WO 2005/017172.

Bharti, A.A.L.N. and Gupta, R.K. (2011) Biotransformation of curcumin to vanillin. *Indian Journal of Chemistry*, **50B**, 1119–1122.

Blom, R.H., Pfeifer, V.F., Moyer, A.J., Traufler, D.H. and Conway, H.F. (1952) Sodium gluconate production – fermentation with *Aspergillus niger*. *Industrial and Engineering Chemistry*, **44**, 435–440.

Boudrant, J. (1990) Microbiol processes for ascorbic acid biosynthesis: a review. *Enzyme and Microbial Technology*, **12**, 322–329.

Boutroux, L. (1980) Chemie physiologique, sur une fermentation nouvelle de glucose (Physiological chemistry, about a new glucose fermentation). *Compte Rendu Academy of Science*, **91**, 236–238.

Breheret, S., Talou, T., Rapior, S. and BessieÁre, J.-M. (1997) Monoterpernes in the aromas of fresh wild mushrooms (*Basidiomycetes*). *Journal of Agricultural and Food Chemistry*, **45**, 831–836.

Bringer-Meyer, S. and Sahm, H. (1991) Process for obtaining sorbitol and gluconic acid by fermentation, and cell material suitable for this purpose. US Patent **5**, 017,485.

Budavari, S., O'Neil, M., Smith, A., Heckelman, P.E. and Kinneary, J.F. (1996) *The Merck Index. An Encyclopedia of Chemicals, Drugs, and Biologicals*. Merck, Whitehouse Station, New Jersey, USA, pp. 1490–1491.

Chalfan, Y., Levy, R. and Mateles, R.I. (1975) Detection of mannitol formation by bacteria. *Applied Microbiology*, **30**, 476.

Chatterjee, T., De, B. and Bhattacharyya, D. (1999) Microbial conversion of isoeugenol to vanillin by *Rhodococcus rhodochrous*. *Indian Journal of Chemistry, Section B – Organic Chemistry Including Medicinal Chemistry*, **38**, 538–541.

Chauhan, A.S., Ramteke, R.S., Eipeson, W.E. (1998) Properties of ascorbic acid and its applications in food processing: a critical appraisal. *Journal of Food Science and Technology*, **35**, 381–392.

Cheethan, P.O.S.J. (1996) Combining the technical push and the business pull for natural flavours. In: Berger, R.G. (ed.), *Biotechnology of Aroma Compounds*. Springer, Berlin and Heidelberg, Germany, pp. 3–49.

Chen, Q., Espey, M.G., Krishna, M.C., Mitchell, J.B., Corpe, C.P., Buettner, G.R., Shacter, E. and Levine, M. (2005) Pharmacologic ascorbic acid concentrations selectively kill cancer cells: action as a pro-drug to deliver hydrogen peroxide to tissues. *Proceedings of the National Academy of Sciences*, **102**, 13604–13609.

Chen, X., Jiang, Z.H, Chen, S. and Qin, W. (2010) Microbial and bioconversion production of D-xylitol and its detection and application. *International Journal of Biological Science*, **6**(7), 834–844.

Cho, Y.K. and Bailey, J.E. (1977) Glucoamylase and glucose oxidase preparations and their combined application for the conversion of maltose to gluconic acid. *Biotechnology and Bioengineering*, **19**, 185–198.

Chun, U.H. and Rogers, P.L. (1988) The simultaneous production of sorbitol and gluconic acid by *Zymomonas mobilis*. *Applied Microbiology and Biotechnology*, **29**, 19–24.

Constaglioli, P., Meilhoc, E., Janatova, I., Ronaldo, D. and Masson, M.J. (1997) Selection of invertase from *Sachwannyomices occidentalis*. *Biotechnology Letters*, **36**(11), 1081–1083.

Converti, A., De Faveri, D., Perego, P., Barghini, P., Ruzzi, M. and Sene, L. (2003) Vanillin production by recombinant strains of *Escherichia coli*. *Brazilian Journal of Microbiology*, **34**(Suppl.), 108–110.

Coutinho Filho, U., Hori, C.E. and Ribiero, E.J. (1999) Influence of reaction products in inversion of sucrose by invertase. *Brazilian Journal of Chemical Engineering*, **16**(2), 149–153.

Elvers, B., Hawkins, S. and Russey, W. (1994) *Ullman's Encyclopedia of Industrial Chemistry*. Wiley-VCH, Weinheim, Germany.

Fabre, C.E., Blanc, P.J. and Goma, G. (1996) Production of benzaldehyde by several strains of *Ischnoderma benzoinum*. *Sciences des Aliments*, **16**, 61–68.

Ferraz, H.C., Alves, T.L.M. and Borges, C.P. (2001) Coupling of an electrodialysis unit to a hollow fiber bioreactor for separation of gluconic acid from sorbitol produced by *Zymomonas mobilis* permeabilized cells. *Journal of Membrane Science*, **191**(1), 43–51.

Ghoreishi, S.M. and Shahrestani, R.G. (2009) Innovative strategies for engineering mannitol production. *Trends in Food Science and Technology*, **20**, 263–270.

Ghosh, S., Sachan, A. and Mitra, A. (2006) Formation of vanillic acid from ferulic acid by *Paecilomyces variotii*. *Current Science*, **90**, 825–829.

Gollhofer, D., Nidetzky, B., Fuerlinger, M. and Kulbe, K.D. (1995) Efficient protection of glucose-fructose oxidoreductase from *Zymomonas mobilis* against irreversible inactivation during its catalytic action. *Enzyme and Microbial Technology*, **17**, 235–240.

Hancock, R.D. and Viola, R. (2005) Improving the nutritional value of crops through enhancement of l-ascorbic acid (vitamin C) content: rationale and biotechnological opportunities. *Journal of Agricultural and Food Chemistry*, **53**, 5248–5257.

Hawkins, S.M. (1993) Bifidobacteria in dairy products. *Cultured Dairy Products Journal*, **28**, 16–20.

Haworth, W.N. and Hirst, E.L. (1933) Synthesis of ascorbic acid. *Journal of the Society of Chemical Industry*, **52**, 645–647.

Herbert, R.W., Hirst, E.L., Percival, E.G.V., Reynolds, R.J.W. and Smith, F. (1933) The constitution of ascorbic acid. *Journal of the Chemical Society*, 1270–1290.

Hirohara, H., Yamamoto, H., Kawano, E. and Nagasa, T. (1982) Continuous hydrolysis of lactose in skim milk and acid whey by immobilized lactase of *Aspergillus oryzae*. In: *Proceedings of the 6th International Enzyme Engineering Conference*, vol. **6**, pp. 295–297.

Honda, Y., Hashiba, H., Ahiko, K.A. and Takahasi, M. (1991) Hydrolysis of immobilized β-galactosidase using a horizontal rotary column reactor. *Nippon Shokulin Kogyo Gakkaish*, **38**, 384.

Husain, Q., Iqbal, J. and Saleemuddin, M. (1985) Entrapment of concanavalin A-glycoenzyme complexes in calcium alginate gels. *Biotechnology Bioengineering*, **27**, 1102–1107.

Jensen, K.A., Evans, K.M.C., Kirk, T.K. and Hammel, K.E. (1994) Biosynthetic pathway for veratryl alcohol in the ligninolytic fungus *Phanerochaete chrysosporium*. *Applied Environmental Microbiology*, **60**, 709–714.

Kastner, G.S. and Egerer, P. (1984) Amino acids and peptides. In: *Biotechnology*, Rehm, H.-J. and Reed, G. (eds), Volume editor K. Kieslich, vol. 6a, *Biotransformation*. Verlag Chemie, Weinheim, Germany, p. 388.

Kawabe, T. and Morita, H. (1994) Production of benzaldehyde and benzyl alcohol by the mushroom *Polyporus tuberaster* K2606. *Journal of Agricultural Food Chemistry*, **42**, 2556–2560.

Keshava, C., Keshava, N., Ong, T. and Nath, J. (1998) Protective effect of vanillin on radiation induced micronuclei and chromosomal aberrations in V79 cells. *Mutation Research*, **397**, 149–159.

Kirk, O., Borchert, T.V. and Fuglsang, C.C. (2002) Industrial enzyme applications. *Current Opinion in Biotechnology*, **13**, 345–351.

Krings, U. and Berger, R.G. (1998) Biotechnological production of flavours and fragrances. *Applied Microbiology*, **49**, 1–8.

Krings, U., Hinz, M. and Berger, R.G. (1996) Degradation of [2H] phenylalanine by the basidiomycete *Ischnoderma benzoinum*. *Journal of Biotechnology*, **51**, 123–129.

Lachke, A.H. and Jeffries, T.W. (1986) Levels of the enzymes of the pentose phosphate pathway in *Pachysolen tannophilus* Y-2460 and selected mutants. *Enzyme and Microbial Technology*, **8**, 353–359.

Lapadatescu, C., Ferron, G., Vergoignan, C., Djian, A., Durand, A. and Bonnarme, P. (1997) Influence of cell immobilization on the production of benzaldehyde and benzyl alcohol by the white-rot fungi *Bjerkandera adusta*, *Ischnoderma benzoinum* and *Dichomitus squalens*. *Applied Microbiology and Biotechnology*, **47**, 708–714.

Lee, W.H. (1967) Carbon balance of a mannitol fermentation and the biosynthetic pathway. *Applied Microbiology*, **15**, 1206–1210.

Leuenberger, H.G.W. (1984) *Methodology in Biotechnology*, vol 6a, *Biotransformations*, Rehm, H.-J. and Reed, G. (eds), Verlag Chemie, Weinheim, Germany, pp. 10–15.

Liese, A., Steelbach, K. and Wandrey, C. (2006) *Industrial Biotransformations*, 2nd edn. Wiley-VCH, New York, USA, and Weinheim, Germany, pp. 273–447.

Lin, J.K. and Lin-Shiau, S.Y. (2001) Mechanisms of cancer chemoprevention by curcumin. *Proceedings of the National Science Council, Republic of China, Part B., Life Sciences*, **25**(2), 59–66.

Lindley, M.G. (1998) The impact of food processing on antioxidants in vegetable oils, fruits and vegetables. *Trends in Food Science and Technology*, **9**, 336–340.

Linko, Y.Y., Weckstro, M.L. and Linko, P. (1980) Sucrose inversion by immobilized *Saccharomyces cerevisiae* yeast cells. In: Food Process Engineering, vol. 2, Linko, P. and Larinkari, J. (eds). Applied Science Publishers Ltd, London, England, pp. 81–91.

Liu, J.Z., Huang, Y.Y., Liu, J., Weng, L.P. and Ji, L.N. (2001) Effects on simultaneous production of glucose oxidase and catalase by *Aspergillus niger*. *Letters in Applied Microbiology*, **32**, 16–19.

Lockwood, L.B. (1979) Organic acid production. In: *The Filamentous Fungi*, vol. 1, Industrial Mycology, Smith, J.E. and Berry, D.R. (eds). Edward Arnold Ltd., London, England, pp. 140–157.

Lupo, M.P. (2001) Antioxidants and vitamins in cosmetics. *Clinics in Dermatology*, **19**, 467–473.

Mabinya, L.V., Mafunga, T. and Brand, J.M. (2006) Determination of ferulic acid and related compounds by thin layer chromatography. *African Journal of Biotechnology*, **5**(13), 1271–1273.

Mahoney, R.R. (1998) Galactosyl-oligosaccharide formation during lactose-hydrolysis: a review. *Food Chemistry*, **63**(2), 147–154.

Marshall, R.O. and Kool, E.R. (1957) Enzymatic conversion of D-glucose to D-fructose. *Science*, **125**, 648–649.

McCaskill, D. and Croteau, R. (1996) Prospects for the bioengineering of isoprenoid biosynthesis. In: *Biotechnology of Aroma Compounds*, Berger, R.G. (ed.). Springer, Berlin, Heidelberg, New York.

Mihalev, K., Schieber, A., Mollov, P. and Carle, R. (2004) Effect of mash maceration on the polyphenolic content and visual quality attributes of cloudy apple juice. *Journal of Agricultural and Food Chemistry*, **52**, 7306–7310.

Molliard, M. (1922) Sur une nouvelle fermentation acide produite par le *Sterigmatocystis nigra*. *Compte Rendu Academy of Science*, **174**, 881–883.

Nidetzky, B., Fürlinger, M., Gollhofer, D., Scopes, R., Haltrich, D. and Kulbe, K. (1997) Improved operational stability of cell free glucose–fructose–oxidorreductase from *Zymomonas mobilis* for the efficient synthesis of sorbitol and gluconic acid in a continuous ultrafiltration membrane reactor. *Biotechnology and Bioengineering*, **53**(6), 623–629.

Panesar, P.S., Kumari, S. and Panesar, R. (2010) Potential application of immobilized β-galactosidase in food processing industries. *Enzyme Research*. DOI: 10.4061/2010/473137.

Parascandola, P. and Scardi, V. (1982) Sucrose inversion by gelatin-entrapped cells of yeast (*Saccharomyces cerevisiae*). *Biotechnology Letters*, **4**, 753–758.

Pazur, J.H. and Kleppe, K. (1964) The oxidation of glucose and related compounds by glucose oxidase from *Aspergillus niger*. *Biochemistry*, **3**, 578–583.

Peppler, H.J. and Reed, G. (1984) Enzymes in food and feed processing. In: *Biotechnology*, vol. 7a, Enzyme Technology, Rehm, H.-J. and Reed, G. (eds). Verlag Chemie, Weinheim, Germany, Pp. 554–556.

Pilizota, V. and Subaric, D. (1998) Control of enzymatic browning of foods. *Food Technology and Biotechnology*, **36**, 219–227.

Ramachandran, S., Fontanille, P., Pandey, A. and Larroche, C. (2006) Gluconic acid: properties, application and microbial production. *Food Technology and Biotechnology*, **44**(2), 185–195.

Rao, R.-S., Jothi, C.P. and Rao, L.V. (2008) Biotechnological production of xylitol by a mutant *Candida tropicalis* OMV5: process optimization using statistical approach. *Indian Journal of Biotechnology*, **7**, 218–224.

Ray, I. and Gupta, M.N. (2003) Lactose hydrolysis by Lactozym™ immobilized on cellulose beads in batch and fluidized bed modes. *Process Biochemistry*, **39**(3), 325–332.

Rebros, M., Rosenberg, M., Mlichova, Z. and Kristofikova, L. (2007) Hydrolysis of sucrose by invertase entrapped in polyvinyl alcohol hydrogel capsules. *Food Chemistry*, **102**, 784–787.

Reichstein, T. and Grussner, A. (1934) Eine ergiebige Synthese der l-Ascorbinsäure (Vitamin C). *Helvetica Chimica Acta*, **17**, 311–328.

Reichstein, T., Grussner, A. and Oppenauer, R. (1933). Die Synthese der d-Ascorbinsäure (d-Form des C-Vitamins). *Helvetica Chimica Acta*, **16**, 561–565.

Ro, H.S. and Kim, H.S. (1991) Continuous production of gluconic acid and sorbitol from sucrose using invertase and an oxidoreductase of *Zymomonas mobilis*. *Enzyme and Microbial Technology*, **13**, 920–924.

Rohr, M., Kubicek, C.P. and Komineck, J. (1983) Gluconic acid. In: *Biotechnology*, vol. 3, Rehm, H.-J. and Reed, G. (eds). Verlag Chemie, Weinhelm, Germany, pp.455–465.

Safarik, I. and Safarikova, M. (1997) *Scientific and Clinical Applications of Magnetic Carriers*, Hafeli, U., Schutt, W., Teller, J. and Zborowski, M. (eds). Plenum Press, New York and London, p. 323.

Safarikova, M. and Safarik, I. (2001) The application of magnetic techniques in biosciences. *Magnetic and Electrical Separation*, **10**, 223–252.

Saha, B.C. (2003) Production of mannitol by fermentation. In: *Fermentation Biotechnology*, Saha, B.C. (ed.). American Chemical Society, Washington, DC, pp. 67–85.

Saha, B.C. (2006) A low-cost medium for mannitol production by *Lactobacillus intermedius* NRRL B-3693. *Applied Microbiology and Biotechnology*, **72**, 676–680.

Saha, B.C. and Racine, F.M. (2008) Production of mannitol by lactic acid bacteria: a review. In: *Biocatalysis and Bioenergy*, Hou, C.T. and Shaw, J.F. (eds). Wiley, Hoboken, USA, pp. 391–404.

Sakurai, H., Lee, H.W., Sato,S., Mukataka, S. and Takahashi, J. (1989) Gluconic acid production at high concentrations by *Aspergillus niger* immobilized on a nonwoven fabric. *Journal of Fermentation and Bioengineering*, **67**, 404–408.

Sankpal, N.V. and Kulkarni, B.D. (2002) Optimization of fermentation conditions for gluconic acid production using *Aspergillus niger* immobilized on cellulose microfibrils. *Process Biochemistry*, **37**, 1343–1350.

Sankpal, N.V., Joshi, A.P., Sutar, I.I. and Kulkarni, B.D. (1999) Continuous production of gluconic acid by *Aspergillus niger* immobilized on cellulosic support: study of low pH fermentative behaviour of *A. niger*. *Process Biochemistry*, **35**, 317–325.

Sauberlich, H.E. (1994) Pharmacology of vitamin C. *Annual Review of Nutrition*, **14**, 371–391.

Scopes, R. K., Rogers, P. L. and Leigh, D. A. (1988) Method for the production of sorbitol and gluconate. US Patent 4755467.

Seitz, E.W. (1994) Fermentation production of pyrazines and terpenoids for flavors and fragrances. In: *Bioprocess Production of Flavor, Fragrance, and Color Ingredients*, Gabelman, A. (ed.). John Wiley & Sons, Inc., New York, USA, pp. 95–134.

Serra, S., Fuganti, C. and Brenna, E. (2005) Biocatalytic preparation of natural flavours and fragrances. *Trends in Biotechnology*, **23**, 193–198.

Silva-Martinez, M., Helitric, D., Novalic, S., Kulbe, K.D. and Nidetzkyb, B. (1998) Simultaneous enzimatic synthesis of gluconic acid and sorbitol: continuous process development using glucose–fructose–oxidorreductase from *Zymomonas mobilis*. *Applied Biochemistry and Biotechnology*, **72**, 863–868.

Silveira, M.M., Lopes da Costa, J.P.C. and Jonas, R. (1994) Processo de produção e recuperação de sorbitol e ácido glucônico ou gluconato. Patente de Invenção PI 9403981-0, INPI, Brazil.

Silveira, M. M., Wisbeck, E., Lemmel, C., Erzinger, G. S., Lopes da Costa, J. P., Bertasso, M. and Jonas, R. (1999) Bioconversion of glucose and fructose to sorbitol and gluconic acid by untreated cells of *Zymomonas mobilis*. *Journal of Biotechnology*, **75**, 99–103.

Simronoff, N. (2001) l-Ascorbic acid biosynthesis. *Vitamins and Hormones*, **61**, 241–266.

Smiley, K.L. and Bolen, K.L. (1982) Demonstration of D-xylose reductase and xylitol dehydrogenase in *Pachysolen tannophylus*. *Biotechnology Letters*, **4**(9), 607–610.

Smiley, K.L., Cadmus, M.C. and Liepins, P. (1967) Biosynthesis of D-mannitol from D-glucose by *Aspergillus candidus*. *Biotechnology Bioengineering*, **9**, 365–374.

Song, H.S. and Vielle, C. (2009) Recent advances in the biological production of mannitol. *Applied Microbiology and Biotechnology*, **84**, 55–62.

Song, K.H., Lee, J.K., Song, J.Y., Hong, S.G., Baek, H., Kim, S.Y. and Hyun, H.H. (2002) Production of mannitol by a novel strain of *Candida magnoliae*. *Biotechnology Letters*, **24**, 9–12.

Svirbely, J.L. and Szent-Gyorgyi, A. (1932) The chemical nature of vitamin C. *Biochemistry Journal*, **26**, 865–870.

Swoboda, B.E.P. and Massey, V. (1965) Purification and properties of the glucose oxidase from *Aspergillus niger*, *Journal of Biological Chemistry*, **240**, 2209–2215.

Szent-Gyorgyi, A.J. (1928) Observations of the function of peroxidase systems and the chemistry of the adrenal cortex: description of a new carbohydrate derivative. *Biochemistry Journal*,**22**, 1387–1409.

Tanriseven, A. and Dogan,S. (2001) Immobilization of invertase within calcium alginate gel capsules. *Process Biochemistry*, **36**(11), 1081–1083.

Tsou, A.Y., Ransom, S.C. and Gerlt, J.A. (1990) Mandelic acid pathway of *Pseudomonas putida*: sequence relationships involving mandelic acid racemase, (*S*)-mandelic acid dehydrogenase, and benzoylformate decarboxylase and expression of benzoylformate decarboxylase in *Escherichia coli*. *Biochemistry*, **29**, 9856–9862.

Vandamme, E.J. and Soetaert, W. (1995) Biotechnical modification of carbohydrates. *FEMS Microbiology Reviews*, **16**, 163–186.

Van der Werf, M.J., De Bont, J.A.M. and Leak, D.J. (1996) Opportunities in microbial biotransformation of monoterpenes. In: *Biotechnology of Aroma Compounds*, Berger R.G. (ed.). Springer, Berlin, Heidelberg, New York.

Waugh, W.A. and King, C.G. (1932) Isolation and identification of vitamin C. *Journal of Biological Chemistry*, **97**, 325–331.

Wisselink, H.W., Weusthuis, R.A., Eggink, G., Hugenholtz, J. and Grobben, G.J. (2002) Mannitol production by lactic acid bacteria: a review. *International Dairy Journal*, **12**, 151–161.

Wrolstad, R.E. and Shallenberger, R.S. (1981) Free sugars and sorbitol in fruits – a compilation from the literature. *Journal of Association of Official Analytical Chemists*, **64**, 91–103.

Zarate, S. and Lopez-Leiva, M.Z. (1990) Oligo-saccharide formation during enzymatic lactose hydrolysis: a literature review. *Journal of Food Protection*, **53**, 262–268.

Znad, H., Markos, J. and Bales, V. (2004) Production of gluconic acid from glucose by *Aspergillus niger*: growth and non growth conditions. *Process Biochemistry*, **39**, 1341–1345.

Section 2
Advanced Processes

17

Ultraviolet in Food Preservation and Processing

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17.1 Introduction

The treatments employed on food mainly aim to obtain products with a longer shelf-life, but especially they must be safe for the consumer. Thermal technologies are the most widely used for these purposes; however, these kinds of treatment have a negative impact on certain components of the food itself, decreasing its vitamin and other nutrient content as well as changing its sensorial characteristics, which make them less attractive concerning their colour and textural properties. Nonthermal technologies are an alternative to the thermal treatments, aiming to obtain a final product with a better sensorial quality without leaving safety aside in terms of its microbial content. These alternative technologies can produce food with no dangerous microorganisms or undesirable enzymes, while maintaining the sensorial and nutritive properties (Noci *et al*., 2008).

Light radiation must be absorbed in order to lead to a photochemical change; thus, the most interesting type of light source to be used will be determined by the absorption spectrum of the reactants. Because of this, to study the radiation effect on a compound, it is necessary to know in which wavelengths the radiation will be absorbed. This spectrum is obtained by irradiating a solution of the compound with known concentrations at

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different wavelengths and measuring the absorbance (Ibarz and Esplugas, 1989; Ibarz, Esplugas and Costa, 1985; Ibarz, Esplugas and Graell, 1985).

In the electromagnetic spectrum, ultraviolet (UV) light fills a wide band of wavelengths in the nonionizing region, with wavelengths between 200 nm (X-rays) and 400 nm (visible light). The UV spectrum can be divided into three regions:

1. Short-length UV light (UV-C) with wavelengths in the interval from 200 to 280 nm.
2. Medium-length UV light (UV-B) with wavelengths in the interval from 280 to 320 nm.
3. Long-length UV light (UV-A) with wavelengths in the interval from 320 to 400 nm.

In addition to having great emission power in the wavelengths in which the reactants absorb, the main characteristics that a radiation source must have in order to be of industrial utility are emission stability, long life, adequate physical dimensions, ease of use and low cost.

The Sun is the most important natural radiation source. Its spectral distribution includes wavelengths from 250 to 1200 nm. Some authors (Bintsis, Litopoulou-Tzanetaki and Robinson, 2000) consider that the flux reaching sea level is from 35 to 50 W/m^2, with an approximate distribution of 9% ultraviolet, 42% visible and 49% infrared radiation. As a consequence, the usable energy fraction to cause chemical changes for UV radiation is very little.

The artificial sources of UV radiation are lamps (Rabek, 1982). There are lamps with a little UV radiation, such as incandescent, halogen and fluorescent lamps, or arches of carbon and plasma. The lamps that irradiate a larger UV fraction are mercury vapour lamps, which consist of a tube filled with mercury in the gaseous state in which an electric current flows.

Concerning the gas pressure, there are low-, medium- and high pressure lamps (Falguera *et al.*, 2011a). The low-pressure lamps emit almost exclusively at 254 nm and as the pressure rises the emission spectrum becomes more complex. It is important to remark that the power of high pressure lamps is much bigger (2500 W) than that of the low-pressure ones (30 W), so that the high pressure lamp irradiates more energy in the desired interval, although it will do the same for other nondesired wavelength intervals.

Other vapour lamps are the sodium ones, with a very high emission fraction in the visible spectrum, and the xenon ones, with a spectral distribution similar to the Sun. The real power of the lamps does not usually match the nominal power, because the lamps lose power over time. To know the real emission power, it is necessary to use actinometries, consisting on well-known photochemical reactions that allow knowing the changes in the concentration of the radiation-sensitive reactant (Rabek, 1982).

There are many types of photochemical reactors (Falguera *et al.*, 2011a, 2011b). They all aim to ensure that all the energy emitted by the lamp reaches, directly or by reflection, the medium where the photochemical reaction will take place. The mathematical model is obtained by performing the mass and energy balances, and the kinetics knowledge (reaction mechanism). Moreover, a radiation balance has to be performed (Bird, Stewart and Lightfoot, 1964) that consists of the evaluation of the total radiation reaching each point and the absorption of this radiation by the medium.

The radiation flow absorbed (W) by the reaction medium can be defined by the expression

$$W_{abs,A} = \int_V \sum_\lambda \epsilon_\lambda I_\lambda C_A \mathrm{d}V \tag{17.1}$$

where ϵ_λ is the molar extinction coefficient of the reaction medium for each wavelength, λ is the wavelength, I_λ is the intensity emited by the lamp for each wavelength, C_A is the reactant concentration and V is the reaction volume.

It is important to define the radiation dose (D). This parameter is obtained by multiplying the radiation flux (D_r, in $\mathrm{W/m^2}$) and the exposure time (t) (Guerrero-Beltrán and Barbosa-Cánovas, 2004; Bintsis, Litopoulou-Tzanetaki and Robinson, 2000). Their units are of energy per area:

$$D = D_r \ t \tag{17.2}$$

When the irradiation is used to inactivate microorganisms, it is usually considered that the kinetics is of the first-order type (Guerrero-Beltrán and Barbosa-Cánovas, 2004), so the number of microorganisms surviving the photochemical treatment can be expressed as

$$N = N_0 \exp(-kD) \tag{17.3}$$

where N and N_0 are the number of microorganisms after the treatment and the initial number, respectively, and k is the rate constant of inactivation. By analogy with conventional thermal treatments, the decimal reduction dose (D_{UV}) is defined as the necessary dose to reduce the number of microorganisms to a tenth:

$$D_{UV} = \frac{1}{k \log e} = \frac{2.303}{k} \tag{17.4}$$

When the treatment takes place in a lamp where the geometry and emission remain unaltered, the decimal reduction dose can be changed by the irradiation time.

One of the biggest limitations presented by UV radiation in the treatment of liquids is its small penetration, which is given by the characteristics of the irradiated liquid. Thus, in distilled water, the loss of radiation intensity at

40 cm from the surface can reach 30%, whereas in a sucrose solution at 10% the same loss of intensity is reached at only 5 cm from the surface (Snowball and Hornsey, 1988). In fruit juices, the ultraviolet light penetration is approximately 1 mm for 90% of the radiation absorption (Sizer and Balasubramaniam, 1999), because of the presence of pigments, pulp content and the presence of melanins and/or melanoidins. This drawback can be solved partially by stirring the reaction medium, thus making sure that all of the molecules of the reactant can reach the irradiated surface (Keyser *et al.*, 2008; Harris and Dranoff, 1965). Falguera *et al.* (2011) studied the radiation absorption in a plane photoreactor with a single lamp as a function of the reaction medium thickness. For a reaction medium with an absorbance of 0.1, hardly 30% of the emitted radiation is absorbed in the first 2.5 cm, while for an absorbance value of 5, 50% of the radiation is absorbed in the first 0.2 cm and nearly the whole radiation is absorbed in the first 1 cm. Fruit juices and other food liquids usually have absorbance values between 2 and 5 for most wavelengths. Thus, although food liquids may have a high absorbance, the radiation does penetrate the liquid.

The most common photochemical reactors are:

- Tubular annular, which consists of a cylinder with an annular section, with the lamp placed in this central annulus space.
- Cylindrical stirred tank, which consists of a perfectly stirred tank with one or more immersed lamps.
- Elliptical reactor, which consists of a cylindrical shell with an elliptical cross-section, constructed with a reflective material to radiation, in which the lamp is placed in a focal axis and the reactor is placed in the other one. Radiant energy emitted by the lamp strikes in the reactor either in a direct way or through the reflection in the cylindrical shell.
- Parallel flat-plate, which consists of two parallel plates placed very close, with the reactants circulating between them, while the radiant energy comes through one side from the outside. This face is made of a material transparent to radiation, which comes either directly from the exterior lamp or by reflection from a parabolic envelope constructed with a reflective material. This kind of reactor is suitable for reactants with very high optical density, since the distance between the plates is short and the fluid flows with a very low thickness, making it easier for the radiation to reach every point in the reactant fluid.
- Descendent film, which consists of a tubular reactor in which the lamp is placed in the central axis and the reactant fluid flows in the form of a film down the inner face of the tube.
- Particle bed, which consists of a bed of glass particles in which there is a layer of a radioisotope, covered with a fluorescent material. High-energy radiation emitted by the radioisotope interacts with the fluorescent material to produce visible or ultraviolet radiation energy.

17.2 Microbial disinfection

Irradiation with ultraviolet light can be used as a disinfection treatment to reduce the microbial load in food (Keyser *et al.*, 2008; Guerrero-Beltrán and Barbosa-Cánovas, 2004; Tran and Farid, 2004). In the food industry, UV-C irradiation is mainly applied for the disinfection of different processes and products, such as air in meat or vegetable processing, water that will be used in later stages of the process, surfaces of fresh products like chicken meat, fish and eggs, and liquid food such as milk, fruit juices or cider (Hadjock, Mittal and Warriner, 2008; Matak *et al.*, 2005; Basaran *et al.*, 2004; Quintero-Ramos *et al.*, 2004; Duffy *et al.*, 2000; Liltved and Landfald, 2000; Wong, Linton and Gerrard, 1998).

UV radiation affects the DNA of the exposed microorganisms. The most effective germicidal wavelengths range between 200 and 280 nm (Tran and Farid, 2004), which matches short wavelength ultraviolet light (UV-C); the maximum germicidal power occurs at 254 nm, whereas its efficiency at 320 nm is almost null (Bintsis, Litopoulou-Tzanetaki and Robinson, 2000). The UV radiation of these wavelengths affects the genetic material of bacteria, virus, mould and other microorganisms, and thus it prevents their reproduction (Hijnen, Beerendonk and Medema, 2006; Billmeyer, 1997; Giese, 1997). The effect of irradiation on the microorganisms depends on various factors such as the species, strain, culture stage and growth phase (Morgan, 1989). Moreover, the type and composition of the food that is irradiated obviously have a great influence.

Table 17.1 shows the maximum and minimum dosages to achieve a total inhibition of different types of microorganisms using a UV-C irradiation source emiting at 254 nm.

17.2.1 Water

The first application of UV irradiation for the disinfection of drinking water was carried out in 1910 in Marseille (Henry, Helbronner and

Table 17.1 Low and high UV-C light dosages (254 nm) needed for complete inhibition of specific microorganisms (source: Guerrero and Barbosa 2004. Reproduced with permission of SAGE.)

Organism	Microorganism	Low dose (J/m^2)	Microorganism	High dose (J/m^2)
Algae	*Chloella vulgaris*	220	Blue green algae	4200
Bacteria (vegetative)	*Bacillus megatherium*	25	*Sarcina lutea*	264
Bacteria (spores)	*Bacillus subtilis*	220	*Bacillus anthracis*	462
Moulds	*Oospora lactis*	110	*Aspergillus niger*	3300
Viruses	Adeno virus types III	45	Tobacco mosaic	4400
Yeasts	Brewer's yeast	66	*Saccharomyces* sp.	176

Recklinghausen, 1910). At that time its use was restricted by the high cost, low reliability of the equipment and the existence of chlorination, which was cheaper, reliable and also allowed to measure the residual disinfectant (Hoyer, 2004; Wolfe, 1990). Since then, UV radiation has gained interest and during the 1980s it has been largely used in Europe for the disinfection of drinking water; in some cases, it has been used to replace chlorination (Gibs, 2000).

The peak use of this technology is because it hardly produces any oxidation-derived subproducts, while chlorination or ozonation do. The true progress in the use of UV irradiation as a primary disinfection process took place after the discovery of its great effectiveness against *Cryptosporidium* (Clancy *et al.*, 1998) and *Giardia*, the two pathogenic microorganisms with a great importance in the safety of drinking water.

There are several works in the literature that studied the disinfection of both drinking and waste water with ultraviolet irradiation (Sommer *et al.*, 2000; Whitby and Palmateer, 1993). Hijnen, Beerendonk and Medema (2006) studied UV irradiation for drinking water disinfection. In the case of waste water disinfection, one of the factors affecting the effectiveness of UV radiation is the quality of the waste water being treated. Thus, UV radiation has shown its effectiveness in the treatment of secondary and tertiary high-quality effluents (Oppenheimer *et al.*, 1997; Blatchley *et al.*, 1996). Nevertheless, such effectiveness becomes more controversial in the case of primary waste waters or low-quality effluents (Whitby and Palmateer, 1993), due to the presence of suspended particles in the waste water, which can absorb a fraction of the imposed radiation, and as a consequence this increases the survival probability of the microorganisms. Whitby and Palmateer (1993) reported the link between suspended solids concentration and the survival rate of faecal coliforms in UV light irradiated waste waters. Taghipour (2004) concluded that in order to reduce the *E. coli* concentration in primary and secondary effluents in one logarithmic cycle, UV radiation doses of 35 and 62 J/m^2, respectively, were needed.

17.2.2 Milk

UV irradiation is also successfully applied for the pasteurization of liquid foods such as milk and fruit juices (Matak *et al.*, 2005; Koutchma *et al.*, 2004). Although the treatment of opaque liquid food with UV irradiation implies an additional problem, this method has been used for different applications in the dairy industry (Bintsis, Litopoulou-Tzanetaki and Robinson, 2000). For example, brines used in the production of Mozzarella cheese have been irradiated (Anon., 1994). Lodi *et al.* (1996) was able to reduce the total colony count by between 50 and 60%, and that of coliforms by 80–90% in goat milk using UV-C radiation. Burton (1951) irradiated pumped milk at high speed through transparent tubes with a 1 cm diameter, so that 80% of the UV radiation reached the milk, destroying approximately 99% of the

bacteria initially present in the milk. Matak *et al.* (2005) demonstrated that UV radiation could be used to reduce the content of *Listeria monocytogenes* in goat milk. For that purpose, fresh goat milk was inoculated with a concentration of 10^7 CFU/mL and irradiated with UV light at doses from 0 to 20 mJ/cm^2; a reduction of the microbial load of more than 5 log cycles was achieved for an accumulative dose of 15.8 mJ/cm^2. In a later work, Matak *et al.* (2007) evaluated the chemical and sensory effects on irradiated goat milk, concluding that an irradiation at 254 nm during 18 s with a dose of 15.8 mJ/cm^2 causes severe sensorial and chemical changes. Milly *et al.* (2007) achieved the inactivation of *E. coli* 25922 in skim milk with a reduction of 3 log cycles. Ibarz, Pagán and Vicente (1986) also reduced the microbial charge in raw cow milk using UV radiation.

Whited *et al.* (2002) studied the effect of light in the vitamin A content on skimmed, semi-skimmed and whole milk, observing that milk fat has a protective effect on the degradation of this vitamin. In all cases, not only the nutritional loss of this vitamin was observed, but also a loss of sensorial quality was noticed.

Furaya, Warthesen and Labuza (1984) studied the photodegradation of riboflavin (vitamin B_2) in macaroni, skimmed milk powder and in buffer solutions. Liquid systems showed first-order photodegradation kinetics and solid food systems a two-step mechanism.

17.2.3 Juices

Juices are among the most sensitive types of food to thermic treatment because they change their colour and lose part of their aroma and vitamins (Choi and Nielsen, 2005). UV radiation is an alternative treatment, since they tend to keep their aroma and colour (Tran and Farid, 2004).

In juices and fruit derivatives, many authors have studied the germicidal effect of UV irradiation on several microorganisms. Gabriel and Nakano (2009) irradiated *Escherichia coli* (K-12 and O157:H7), *Salmonella* (*enteriditis* and *typhimurium*) and *Listeria monocytogenes* (AS-1 and M24-1) strains in phosphate buffer solution and in clarified apple juice. S. *typhimurium* came to be the most sensitive to ultraviolet radiation with a decimal reduction time of 0.27 min, whereas *Listeria monocytogenes* AS-1 came to be the most resistant with a value of 1.26 min.

Keyser *et al.* (2008) successfully used UV irradiation to reduce the microbial load in different fruit juices and nectars. In clarified apple juice, they managed to reduce in more than 7 log cycles the *E. coli* population with a dose of 1377 J/L, whereas a 230 J/L dose sufficed to reduce the count of aerobic mesophyll microorganisms in 3.5 log cycles and 3 log cycles for moulds and yeasts. In orange juice with a cell content between 7.5 and 10% in weight and after a radiation dose of 1607 J/L, reductions of only 0.3 log cycles for aerobic mesophylls, moulds and yeasts were obtained. This small efficiency is caused by the great quantity of suspended matter, such as orange

cells and fibre present in orange juice, which acts as a protective barrier for microorganisms against UV radiation.

Guerrero-Beltrán and Barbosa-Cánovas (2005) studied the decrease of *Saccharomyces cerevisiae*, *Escherichia coli* and *Listeria innocua* populations in apple juice by UV radiation. The obtained results showed that as the treatment time and flow rate increased, there was a greater probability that these microorganisms were damaged or inactivated by the radiation. Walkling-Ribeiro *et al.* (2008) treated apple juice with previously inoculated *Staphylococcus aureus* (SST 2.4) by a combined UV radiation, pre-heating and high-density electrical pulse method, reducing the microbial population by up to 9.5 log cycles. In the most severe conditions, the reduction achieved using this combined method became even higher than that of conventional pasteurization. Ngadi, Smith and Cayoutte (2003) reduced the count of *E. coli* O157:H7 in apple juice by about 4.5 log cycles using a dose of 3000 mJ/cm^2 and a liquid depth of 1 mm. This type of treatment has also been used to reduce the microbial load in apple cider (Wright *et al.*, 2000). Worobo (1999) also managed to reduce the *E. coli* population for apple cider by more than 5 log cycles. Milly *et al.* (2007) achieved the inactivation of *E. coli* 25922 in apple juice with a reduction of 4.5 log cycles.

Guerrero-Beltrán *et al.* (2009) processed grape, cranberry and grapefruit pasteurized juices inoculated with *S. cerevisiae*, using an UV-C disinfection unit and working at different flow rates and doses of UV light (75–450 kJ/m^2). The inactivation followed a first-order kinetics with a decimal reduction time ranging from 61.7 to 113.7, 12.2 to 40.7 and 12.5 to 20.7 min for grape, cranberry and grapefruit juices, respectively. The maximum reduction log was 0.53, 2.51 and 2.42 for yeast count in grape, cranberry and grapefruit juices, respectively, after 30 min of treatment.

Pesek and Warthesen (1990) studied the photodegradation kinetics of β-carotene by irradiating model solutions and carrot juices, and reported first-order kinetics; the cis isomer was more sensitive to photodegradation. Pesek and Warthesen (1987) studied the effects of irradiation on lycopene, α- and β-carotene in tomato and carrot juices; first-order kinetics with kinetic constants from 0.1 to 0.3 per day were reported and lycopene was the most resistant compound to photodegradation.

Falguera, Pagán and Ibarz (2011) observed a loss between 4 and 6% of vitamin C in 120 min of irradiation of apple juices from the varieties Golden, Starking and Fuji, whereas the Kind David variety showed a loss of 70%. The difference was attributed to the lack of pigmentation of the Kind David variety juice. Tran and Farid (2004) found a vitamin C degradation of 12% in orange juice with a UV dose of 73.8 mJ/cm^2 at 254 nm.

UV radiation also allows the degradation of pesticide remains coming from fruit, such as pyridine (Ibarz, Esplugas and Costa, 1985), carbendazim (Ibarz and Pérez-Teijón, 1990), benomyl (Ibarz, Panadés and Tejero, 1996), thiabendazole (Panadés, Alonso and Ibarz, 1997) and indole (Ibarz *et al.*, 1998).

Attoe and von Elbe (1981) studied the effect of light on betanine extracted from beet and on cranberry anthocyanins. For both types of pigments, it was found that photochemical degradation follows first-order kinetics; the presence of molecular oxygen is of importance in the degradation of these pigments.

17.2.4 Beers and other drinks

In brewing and beverage industries, many manufacturers have adopted UV radiation as a disinfection system for the water used in the process, since it is essential that the applied treatment does not affect the taste and quality of the final product (Greig and Warne, 1992). The UV doses required for water treatment in the brewing industry are much higher than those used in water purification treatments, since it must guarantee the absence of microbes during the first stages of beer manufacturing.

Lu *et al.* (2010) achieved a reduction of inoculated *S. cerevisiae* and *L. brevis* in beer of around 5 log cycles and from 10^4 CFU/mL to nondetectable limits at doses of 16.1 and 9.7 mJ/cm^2, respectively. However, the beneficial yeasts of beer were hardly inactivated.

17.2.5 Liquid egg derivatives

In liquid egg derivatives, the UV-C irradiation can become an alternative treatment to obtain a microbiologically safe and stable product (Donahue, Canitez and Bushway, 2004; Bintsis, Litopoulou-Tzanetaki and Robinson, 2000), while avoiding the alterations that other methods such as high hydrostatic pressures, high-intensity pulsed electric fields or pasteurization cause in the product because of protein denaturation (Unluturk *et al.*, 2008).

Ngadi, Smith and Cayoutte (2003) studied the UV irradiation of liquid egg white (pH 9.1) inoculated with *E. coli* O157:H7; a decrease in the microorganisms count from 10^8 to $10^{3.8}$ CFU m/L was obtained after being exposed to a 300 mJ/cm^2 dose. Unluturk *et al.* (2008) studied the effect of UV irradiation on nonpathogenic *E. coli* (ATCC 8739) and *Salmonella typhimurium* strains in liquid egg derivatives. The maximum reduction, more than 2 log cycles, was obtained for *E. coli* (ATCC 8739) in liquid egg white with a liquid depth of 0.153 cm and UV radiation flux of 1314 mW/cm^2. Nevertheless, under the same conditions for liquid egg yolk and liquid whole egg, the maximum reductions obtained were only 0.675 and 0.361 log cycles, respectively.

17.2.6 Food surfaces and packaging

One of the main industrial applications of ultraviolet radiation is the sterilization of packaging materials such as containers, wrapping or bottle caps

(Bintsis, Litopoulou-Tzanetaki and Robinson, 2000). In the aseptic packaging of UHT-treated products such as milk, the UV radiation is used to sterilize the aluminum bottle caps (Nicolas, 1995) or carton containers (Kuse, 1982). The materials for the aseptic processing and packaging can also be sterilized by combining the treatments with hydrogen peroxide and ultraviolet radiation (Marquis and Baldeck, 2007), taking advantage of its synergistic effect on the destruction of bacterial spores. The combined use of ozone and UV radiation for the treatment of plastic polymers used for food packaging has also been studied (Ozen and Floros, 2001).

UV-C radiation can also be used for the treatment of food surfaces. Thus, there are several studies that demonstrate the efficiency of UV radiation to reduce the surface population of pathogenic microorganisms in red meat, chicken and fish (Sumner *et al.*, 1995). Wong, Linton and Gerrard (1998) demonstrated the effectiveness of UV light to reduce the content on *E. coli and S. senftenberg* on pig skin and muscle surfaces. The UV light was more effective on *S. senftenberg* than on *E. coli* and faster on pig skin than on muscle. Dejenane *et al.* (2001) found that the commercial life of fresh meat can rise from 12 to 28 days due to UV treatment. The *S. typhimurium* content was reduced by UV treatment in a chicken skeleton without affecting its colour (Wallner-Pendleton *et al.*, 1994). Lyon, Fletcher and Berrang (2007) reduced 2 log cycles of the *L. monocytogenes* concentration on chicken breast fillets with an UV light treatment. Chun *et al.* (2010) demonstrated that UV-C irradiation treatments allow the microbial load of *Campylobacter jejuni*, *L. monocytogenes* and *S. typhimurium* in chicken breast to be reduced. Likewise, UV-C radiation can be used as a method to improve the microbial safety of ready-to-eat food such as ham slices (Chun *et al.*, 2009).

Kuo, Carey and Ricke (1997) demonstrated that UV-C radiation becomes effective to reduce the count of total aerobics and moulds, as well as *S. typhimurium* on egg shells. Other studies demonstrated the effectiveness of UV-C to reduce the surface microbial load on vegetables (Allende and Artés, 2003) and fruits (González-Aguilar *et al.*, 2001). UV-C radiation has also been used to reduce post-harvest deterioration of onion (Lu *et al.*, 1987), carrot (Mercier and Arul, 1993), tomatoes (Maharaj, 1995) and zucchini (Erkan, Wang and Krizek, 2001).

There are also studies confirming the effectiveness of UV-C radiation to reduce diseases in fruits such as peach and apple (Stevens *et al.*, 1996), table grapes (Nigro, Ippolitto and Lima, 1998), grapefruit (Droby *et al.*, 1993) and papaya (Cia *et al.*, 2007).

UV-C radiation can also be used to treat fresh fruits, vegetables and edible roots before their packaging. The beneficial effect of this kind of treatment on fresh food is called 'hormesis' (Stevens *et al.*, 1999, 1997). This effect is caused because the UV light can stimulate the production of phenylalanine amonialyase (PAL), which leads to the formation of phenolic compounds (phytoalexins); these compounds can improve the resistance of fruit and

vegetables against microorganisms (D'Halewin *et al*., 2000; Stevens *et al*., 1999). Onursal *et al.* (2010) found that the irradiation of pomegranate fruits increased the total phenolics content in juice, peel and seeds. The irradiation of broccoli was shown to delay senescence and to increase the antioxidant capacity (Costa *et al*., 2005).

17.2.7 Air disinfection

The use of UV-C radiation as a germicidal agent for air decontamination is a known method from decades ago. The microorganisms are more sensitive to UV-C if they are suspended in water and these, in turn, are more sensitive to those found in fruit juices (Bintsis, Litopoulou-Tzanetaki and Robinson, 2000). Jensen (1964) irradiated aerosolized viruses by making them pass through an aluminium cylindrical tube with a highly reflective internal surface at the centre of which the UV lamp was placed, reaching (in the most favourable conditions) an inactivation higher than 99.9% for *Coxsackie*, *Influenza*, *Sindbis* and *Vaccinia* viruses. Xu *et al.* (2003) evaluated the germicidal effectiveness of the UV radiation on bacterial spores and vegetative mycobacterial cells and obtained a reduction of between 46 and 80% for *Bacillus subtilis* spores and between 83 and 98% for *Mycobacterim parafortuitum* ones.

Josset *et al.* (2007) designed a new photoreactor for air decontamination at high airflow using UV-A radiation. Inactivation rates of 93% were obtained in a single pass through the photoreactor with airflow of 5 m^3/h and with a concentration of 1.2×10^6 CFU/L of *Legionella pneumophila*.

17.3 Mycotoxin elimination

The presence of mycotoxins involves a severe problem for food safety. In conventional thermal processes, mycotoxins are not affected, and thus it becomes essential to find an alternative treatment to eliminate or at least reduce its content in food.

Leeson, Díaz and Summers (1995) demonstrated that it is possible to destroy aflatoxins in peanuts, using UV radiation and sunlight. In the case of citrinin and ochratoxin A, Neely and West (1972) found that there is a limited amount of decomposition when treated with UV light.

UV radiation has been used to degrade aflatoxin M_1 in raw and heated milk (Yousef and Marth, 1985); if irradiation is carried out at 25 °C, the reduction of aflatoxin M_1 is 32% higher than when it takes place at 5 °C. Nevertheless, when the treatment temperature is 65 °C, the reduction is only 25.5%. These data suggest that it is possible to degrade milk aflatoxins at a lower temperature than pasteurization. Yousef and Marth (1987) studied the degradation

of aflatoxin M_1 in aqueous solution and observed that its elimination was accompanied by an accumulation of aflatoxin M_x, which was also degraded when the treatment temperature was increased to 60 °C.

Peanut oil is the most common food containing aflatoxins. Shantha and Sreenivasa Murthy (1977) studied the effect of light of different wavelengths, using lamps that emit in the UV and Sun regions. The results show that for long irradiation times, high elimination rates can be obtained; 87% of destruction with UV and 82% with sunlight have been reported. The absorption spectra of the samples containing aflatoxin after being exposed to UV and sunlight show a shift of the absorption maximum that is accompanied by a significant reduction in toxicity. Nkama and Muller (1988) observed that aflatoxin B_1 contained in rice is degraded by the action of light emitted by a mercury-tungsten lamp. Samples containing approximately 1000 mg/kg showed a decrease of 70% after 2 h of irradiation at 64 mW/cm^2 and 60% with 43 mW/cm^2.

Samarajeewa *et al.* (1990) made a review of physical and chemical methods used in aflatoxin detoxification for food and animal feed. In particular, they highlighted the effect of ultraviolet and visible light. Aflatoxin B_1 has an absorption spectrum with peaks at 222, 265 and 362 nm wavelengths, being the maximum 362 nm. Irradiation at this wavelength excites aflatoxin and rises its susceptibility to degradation, being highly sensitive to UV radiation at pH values below 3 or above 10, since the aflatoxin structure is affected in the terminal furan ring, eliminating the active point of link (Samarajeewa *et al.*, 1990).

Solar radiation, which contains radiant energy in the ultraviolet and visible spectra, has proved a great efficacy in the degradation of aflatoxins in food (Samarajeewa and Arseculeratne, 1974). Kinetic studies of aflatoxin degradation show first-order kinetics and that the intermediate products formed have some toxicity (Aibara and Yamagishi, 1968). To lead the aflatoxin breakdown towards nontoxic secondary compounds, a white light source can be used (Samarajeewa *et al.*, 1990). Edible oils contaminated with aflatoxin and irradiated with sunlight have not shown any toxicity in tests with mice and ducks (Samarajeewa, Gamage and Arseculeratne, 1987; Shantha and Sreenivasa Murthy, 1980, 1977). Degradation of aflatoxin B_1 with sunlight in food suggests the presence of aflatoxin in two states, one accessible and the other less accessible. The accessible aflatoxin is easily degraded, being described as 'unbound' in casein and peanut (Shantha and Sreenivasa Murthy, 1981), following the first-order kinetics during its degradation in rice (Nkama, Mobbs and Muller, 1987). The less accessible aflatoxin is described as 'linked' and does not follow first-order kinetics during its degradation. These differences are probably associated with the fact that aflatoxin on the surface of food is accessible to solar radiation, whereas those inside the food are protected by the low penetration of this type of radiation.

In the case of liquid foods, solar radiation can penetrate more easily so that the radiation has a higher decontamination capacity. Studies carried out at a pilot plant scale have showed the efficiency of solar radiation on aflatoxin degradation in coconut and peanut oil (Samarajeewa *et al.*, 1985; Shantha and Sreenivasa Murthy, 1981).

17.4 Inactivation of enzymes in juices

Enzymes play an important role in the manufacture of fruit derived juices, whether they are clarified or purees. Polyphenol oxidase (PPO) is the enzyme causing the enzymatic browning by oxidation of the colourless phenolic groups to coloured ketonic groups that end up forming melanins by polymerization. Therefore, it is important to inactivate this enzyme to avoid this undesirable colour deterioration in juices. A studied method is UV irradiation, achieving not only the inactivation of PPO but also inhibiting the discoloration through pigments and melanins degradation (Falguera, Pagán and Ibarz, 2011; Guerrero-Beltrán and Barbosa-Cánovas, 2005; Ibarz and Pérez-Teijón, 1990). Seiji and Iwashita (1965) found that melanins themselves have a protective effect against impairment. Another similar degradation is the nonenzymatic browning that produces coloured melanoidins by the Maillard reaction between reducing sugars and the carbonyl groups of free amino acids; this reaction is sensitive to temperature. Once it has occurred, one of the ways to eliminate melanoidins is by UV irradiation (Ibarz *et al.*, 2005; Kwak *et al*., 2004).

Other enzymes of great importance in juices are pectic enzymes, which cause pectin degradation; examples are pectinmethylesterase (PME), endo-polygalacturonase (endo-PG) and exo-polygalacturonase (exo-PG). The inactivation of pectic enzymes is essential in fruit derivatives with suspended pulp to avoid sedimentation of the suspended solid fraction.

Tran and Farid (2004) irradiated orange juice with UV at 254 nm and a dose of 73.8 mJ/cm^2, obtaining a pectinmethylesterase inactivation of 5%. The juice contained suspended pulp, which is known for the protective effect against light. It is likely that it would have obtained a higher extent of inactivation if a lamp with a wider emission spectrum had been used, as Falguera, Pagán and Ibarz (2011) did, managing to completely inactivate polyphenol oxidase after 100 min and peroxidase after 15 min in the irradiation of four different varieties of apple juices with a mercury vapour lamp of 400 W medium pressure with an emission interval between 250 and 650 nm. The same lamp was used by Ibarz *et al.* (2009) to completely inactivate the gastric enzymes, such as carboxypeptidase A and trypsin.

The inactivation of the enzymes chymotrypsin, lysozyme, ribonuclease and trypsin with ultraviolet light at 253.7 nm (Luse and McLaren, 1963)

was attributed to the absorption by a specific amino acid residue. McLaren *et al.* (1953) inactivated ribonuclease and carboxypeptidase using UV light (253.7 nm), obtaining quantum yields of 0.03 and between 0.001 and 0.005, respectively. The quantum yields for low molecular weight protein are of the order of 0.03 and are higher than those of the peptide bonds; it is postulated that the primary inactivation process carries the modification of the aromatic residues in proteins.

In addition, tyrosinase, acid phosphatase and adenosine triphosphatase can be denatured when irradiated with ultraviolet light, with the resultant decrease in enzymatic activity (Seiji and Iwashita, 1965). The loss of enzymatic activity depends on the radiation intensity, with the possibility of reaching inactivation degrees of 75% for tyrosinase and around 50% for the other two enzymes.

17.5 Improvement of polymer films

Bipolymer films have recently been applied in the food industry as an alternative to nonbiodegradable plastic films. UV radiation treatments can improve the properties of bipolymer films because double bonds and aromatic rings can absorb this type of radiation, causing the formation of free radicals in amino acids, which leads to the subsequent formation of intermolecular covalent bonds (Wihodo and Moraru, 2013; Rhim *et al*., 1999; Gennadios *et al*., 1998).

The films that are formed from proteins show a different response to UV irradiation (253.7 nm) because of the variation of the molecular structures and the composition of the amino acids that form them. Gennadios *et al.* (1998) treated soy protein films with UV radiation at different light intensities (13 to 103.7 J/m^2), observing a linear rise of the tensile strength with an increase in the radiation dose, whereas the elongation at break decreased linearly with the dose. Moreover, the permeability of films against water vapour was not affected. Likewise, the SDS-PAGE bands of the irradiated samples showed aggregate bands, which increased with the treatment dose. The soy protein contains significant amounts of tyrosine and phenylalanine, which are aromatic compounds that can absorb UV radiation and recombine to form covalent cross-links. UV irradiated films developed a yellow colouration, with a linear increase of the colourimetric parameter *b* (Hunter b value) with an increase in the radiation dose; moreover, luminosity *L* (Hunter L value) and parameter *a* (Hunter a value) showed a slight variation, tending towards darker and greener values.

Rhim *et al.* (1999) treated films formed from wheat gluten, corn zein, egg albumin and sodium caseinate with UV radiation at a dose of 51.8 J/m^2. The UV-treated films of gluten, zein and albumin showed an increase in tensile

strength. The UV radiation reduced the permeability of water vapour to albumin films, but other films were not affected. Concerning colorimetric parameters, luminosity L and the parameter a barely had any variation; nevertheless, the paramater b showed an increase in the irradiated films (yellower), with the exception of the corn zein sample, probably because the UV radiation destroyed the pigments of the sample itself.

Wheat gluten films treated with UV radiation, at dosages of 0.25 and 1 J/cm^2, showed little variation in tensile strength values, elongation and Young's modulus (Micard *et al.*, 2000). Likewise, Gueguen *et al.* (1998) treated pea films previously plasticized with ethylene glycol and hydroxyethylacrylate, obtaining a slight increase in the tensile strength after UV radiation at 0.5 J/cm^2.

Shi, Kokini and Huang (2009) showed that UV/ozone treatment of zein films can be used to obtain films with controlled hydrophilicity. This treatment is able to cause the oxidation of superficial groups through a sequence of reactions in which UV radiation can excite molecular oxygen to form atomic oxygen and ozone, which are highly reactive so that hydroxyl radicals are generated. Likewise, UV radiation can excite the zein film forming free radicals. All these radical-forming species react with the film surface and can oxidize it, turning some ethyl groups into carbonyl groups, thereby modifying the hydrophilicity of the film.

Solutions of whey protein isolate, irradiated at 324 J/cm^2 (Ustunol and Mert, 2004), obtained films with a tensile strength slightly higher than the control film; however, the elongation was lower. On the other hand, permeability of water vapour showed a slight increase, whereas permeability slightly decreased.

17.6 Conclusions

Ultraviolet irradiation constitutes an alternative to thermal treatment that is being studied and developed to obtain a better final product sensory quality considering microbial safety. For each application, it is necessary to select the radiation source, reactor geometry and reaction medium properties. The absorbed amount of radiation has a definitive effect on the reaction rate of the process in each point of the reaction medium; thus, it is essential to know the radiation model with an overall intention of applying it in the area of food preservation and processing.

References

Aibara, K. and Yamagishi, S. (1968) Effects of ultra-violet radiation on the destruction of aflatoxin B1. In: *Proceedings of the first US–Japan Conference on Toxic Microorganisms*, UJNT Joint Panel on Toxic Microorganisms and US Department of Interior, Washington, DC.

Allende, A. and Artés, F. (2003) UV-C radiation as a novel technique for keeping quality of fresh processed 'Lollo Rosso' lettuce. *Food Research International*, **36**, 739–746.

Anon. (1994) Mozzarella cheese protected by ultraviolet disinfection. *Food Industry*, **47**(10), 19–21.

Attoe, E.L. and von Elbe, J.H. (1981) Photochemical degradation of betanine and selected anthocyanins. *Journal of Food Science*, **46,** 1934–1937.

Basaran, N., Quintero-Ramos, A., Moake, M.M., Churey, J.J. and Worobo, R.W. (2004) Influence of apple cultivars on inactivation of different strains of *Escherichia coli* O157:H7 in apple cider by UV irradiation. *Applied Environmental Microbiology*, **70**, 6061–6065.

Billmeyer, F.W. (1997) Ultraviolet lamp. In: *Encyclopedia of Science and Technology*, 8th edn, vol. **19**. McGraw-Hill, New York, USA, pp. 19–20.

Bintsis, T., Litopoulou-Tzanetaki, E. and Robinson, R. (2000) Existing and potential applications of ultraviolet light in the food industry – a critical review. *Journal of the Science of Food and Agriculture*, **80**, 637–645.

Bird, R., Stewart, W. and Lightfoot, E. (1964) *Fenómenos de Transporte*. Ed. Reverté, Barcelona, Spain.

Blatchley, E.R., Bastian, K.C., Duggirala, R.K., Alleman, J.E., Moore, M. and Schuerch, P. (1996) Ultraviolet irradiation and chlorination/dechlorination for municipal wastewater disinfection. *Water Environmental Research*, **68**, 194–204.

Burton, H. (1951) Ultra-violet irradiation of milk. *Dairy Science Abstracts*, **13**, 229–244

Choi, L.H. and Nielsen, S.S. (2005) The effect of thermal and non-thermal processing methods on apple cider quality and consumer acceptability. *Journal of Food Quality*, **28**, 13–29.

Chun, H.H., Kim, J.Y., Chung, K., Won, M. and Song, K.B. (2009) Inactivation kinetics of *Listeria monocytogenes*, *Salmonella enteric typhimurium* and *Campylobacter jejuni* in ready to eat sliced ham using UV-C irradiation. *Meat Science*, **83**, 599–603.

Chun, H.H., Kim, J.Y., Lee, B.D., Yu, D.J. and Song, K.B. (2010) Effect of UV-C irradiation on the inactivation of inoculated pathogens and quality of chicken breast during storage. *Food Control,* **21***,* 276–280.

Cia, P., Pascholati, S.F., Benato, E.A., Camili, E.C. and Santos, C.A. (2007) Effects of gamma and UV-C irradiation on the postharvest control of papaya antracnose. *Postharvest Biology and Technology*, **43**, 366–373.

Clancy, J.L., Hargy, T.M., Marshall, M.M. and Dyksen, J.E. (1998) UV light inactivation of *Cryptosporidium oocysts*. *Journal of the American Water Works Association*, **90**, 92–102.

Costa, L., Vicente, A.R., Civello, P.M., Chaves, A.R. and Martínez, G.A. (2005) UV-C treatment delays post harvest senescence in broccoli florets. *Postharvest Biological Technology*, **39**, 204–210.

Dejenane, D., Sánchez-Escalante, A., Beltrán, J.A. and Roncalés, P. (2001) Extension of the retail display life of fresh beef packaging in modified atmosphere by varying lighting conditions. *Journal of Food Science*, **66**(1), 181–186.

D'Hallewin, G., Schirra, M., Pala, M. and Ben-Yehoshua, S. (2000) Ultraviolet C irradiation at 0.5 kJ m^{-2} reduces decay without causing damage or affecting post harvest quality of Star Ruby grapefruit (*C. paradise* Macf.). *Journal of Agriculture and Food Chemistry*, **48**, 4571–4575.

Donahue, D.W., Canitez, N. and Bushway, A.A. (2004) UV inactivation of *E. coli* 0157:H7 in apple cider: quality, sensory and shelf life analysis. *Journal of Food Processing and Preservation*, **28**, 368–387.

Droby, S., Chalutz, E., Horev, B., Cohen, L., Gaba, V., Wilson, C.L. and Wisniewski, M.E. (1993) Factors affecting UV-induced resistance in grapefruit against the green mold decay caused by *Penicillium digitatum*. *Plant Pathology*, **42**, 418–424.

Duffy, S., Churey, J.J., Worobo, R.W. and Schaffner, D.W. (2000) Analysis and modelling of the variability associated with UV inactivation of *Escherichia coli* in apple cider. *Journal of Food Protection*, **63**, 1587–1590.

Erkan, M., Wang, C.Y. and Krizek, D.T. (2001) UV-C radiation reduces microbial populations and deterioration in *Cucurbita pepo* fruit tissue. *Environmental andExperimental Botany*, **45**, 1–9.

Falguera, V., Pagán, J., Ibarz, A. (2011) Effect of UV irradiation on enzymatic activities and physicochemical properties of apple juices from different varieties. *LWT-Food Science and Technology*, **44**(1), 115–119.

Falguera, V., Esplugas, S., Vicente, M. and Ibarz, A. (2011a) Modeling of absorbed radiation profiles in a system composed by a plane photoreactor and a simple lamp. *Food Research International*, **44**, 3111–3114.

Falguera, V., Pagán, J., Garza, S., Garvín, A. and Ibarz, A. (2011b) Ultraviolet processing of liquid food: a review. Part 1: Fundamental engineering aspects. *Food Research International*, **44**, 1571–1579.

Falguera, V., Pagán, J., Garza, S., Garvín, A. and Ibarz, A. (2011c) Ultraviolet processing of liquid food: A review. Part 2: Effects on microorganisms and on food components and properties. *Food Research International*, **44**, 1580–1588.

Furaya, E.M., Warthesen, J.J. and Labuza, T.P. (1984) Effects of water activity, light intensity and physical structure on the kinetics of riboflavine photo degradation. *J. Food Science*, **49**, 525–528.

Gabriel, A.A. and Nakano, H. (2009) Inactivation of *Salmonella*, *E. coli* and *Listeria monocytogenes* in phosphate-buffered saline and apple juice by ultraviolet and heat treatments. *Food Control*, **20**, 443–446.

Gennadios, A., Rhim, J.W., Handa, A., Weller, C.L. and Hanna, M.A. (1998) Ultraviolet radiation affects physical and molecular properties of soy protein films. *J. Food Science*, **63**(2), 225–228.

Gibbs, C. (2000) UV disinfection. *Soft Drink International*, January, 32–34.

Giese, A.C. (1997) Ultraviolet radiation (biology). In: *Encyclopedia of Science and Technology*, 8th edn, vol. 19. McGraw-Hill, New York, USA, pp. 20–22.

González-Aguilar, G.A., Wang, C.Y., Buta, J.G. and Krizek, D.T. (2001) Use of UV-C irradiation to prevent decay and maintain post harvest quality of ripe 'Tommy Atkins' mangoes. *International Journal of Food Science and Technology*, **36**, 767–773.

Greig, C. and Warne, S. (1992) UV disinfection systems aid brewery hygiene. *Food Technology*, **27**(3), 20–21.

Gueguen, J., Viroben, G., Noireaux, P. and Subirade, M. (1998) Influence of plasticizers and treatments on the properties of films from pea proteins. *Industrial Crops Production*, **7**, 149–157.

Guerrero-Beltrán, J.A. and Barbosa-Cánovas, G.V. (2004) Advantages and limitations on processing foods by UV Light. *Food Science and Technology International*, **10**, 137–147.

Guerrero-Beltrán, J.A. and Barbosa-Cánovas, G.V. (2005) Reduction of *Saccharomyces cerevisae*, *Escherichia coli*, *and Listeria innocua* in apple juice by ultraviolet light. *Journal of Food Process Engineering*, **28**, 437–452.

Guerrero-Beltrán, J.A., Welti-Chanes, J. and Barbosa-Cánovas, G.V. (2009) Ultraviolet-C light processing of grape, cranberry and grapefruit to inactivate *Saccharomyces cervisae*. *Journal of Food Process Engineering*, **32**, 916–932.

Hadjock, C., Mittal, G.S. and Warriner, K. (2008) Inactivation of human pathogens and spoilage bacteria on the surface and internalized within fresh produce by using a combination of ultraviolet light and hydrogen peroxide. *Journal of Applied Microbiology*, **104**, 1014–1024.

Harris, P.R. and Dranoff, J.S. (1965) A study of perfectly mixed photochemical reactors. *AIChE Journal*, **11**, 497–502.

Henry, V., Helbronner, A. and Recklinghausen, M. (1910) Nouvelles recherches sur la sterilization de grandes quantités d'eau par les rayons ultraviolets. *Comptes Rendus de l'Académia de Sciences*, **151**, 677–680.

Hijnen, W.A.M., Beerendonk, E.F. and Medema, G.J. (2006) Inactivation credit of UV radiation for viruses, bacteria and protozoan (oo) cysts in water: a review. *Water Research*, **40**, 3–22.

Hoyer, O. (2004) Water disinfection with UV radiation – requirements and realization. In: *Proceedings of the European Conference UV Karlsruhe, UV radiation. Effects and Technologies*, September 2003, Karlsruhe, pp. 22–24.

Ibarz, A. and Esplugas, S. (1989) Ingeniería fotoquímica. Aplicación a la industria alimentaria. *Theknos*, **110**, 8–14.

Ibarz, A., Esplugas, S. and Costa, J. (1985a) Influencia del pH en la fotodescomposición de piridina. *Afinidad*, **397**, 265–269.

Ibarz, A., Esplugas, S. and Graell, J. (1985b) Procesos fotoquímicos: aplicación a la fotodescomposición de contaminantes en agua y alimentos. *Alimentación; Equipos y Tecnología*, **3**(85), 155–164.

Ibarz, A., Garvín, A., Garza, S. and Pagán, J. (2009) Inactivation of carboxypeptidase A and trypsin by UV-Visible light. *Innovative Food Science and Emerging Technologies*, **10**(4), 517–521.

Ibarz, A., Pagán, J. and Vicente, M. (1986) Efecto de la radiación ultravioleta sobre leche cruda. *Alimentación; Equipos y Tecnología*, **4**(86), 95–101.

Ibarz, A., Panadés, R. and Tejero, J.M. (1996) Fotodescomposición de benomilo en solución acuosa. *Afinidad*, **461**, 53–56.

Ibarz, A. and Pérez-Teijón, C.J. (1990) Efecto de la radiación solar sobre zumo de manzana y carbendazima en solución acuosa. *Alimentaria*, **214**, 33–37.

Ibarz, A., Tejero, J.M., Panadés, R., Esplugas, S. (1998) Descomposición fotoquímica de indol en solución acuosa. *Tecnología del Agua*, **173**, 47–60.

Ibarz, A., Pagán, J., Panadés, R. and Garza, S. (2005) Photochemical destruction of color compounds in fruit juices. *Journal of Food Engineering*, **69**(2), 155–160.

Jensen, M.M. (1964) Inactivation of airborne viruses by ultraviolet irradiation. *Applied Environmental Microbiology*, **12**, 418–420.

Josset, S., Taranto, J., Keller, N., Keller, V., Lett, M.C., Ledoux, M. J., Bonnet, V. and Rougeau, S. (2007) UV-A photocatalytic treatment of high flow rate air contaminated with *Legionella pneumophila*. *Catalysis Today*, **129**, 215–222.

Keyser, M., Müller, I.A., Cilliers, F.P., Nel, W. and Gouws, P.A. (2008) Ultraviolet radiation as non-thermal treatment for inactivation of microorganisms in fruit juice. *Innovative Food Science and Emerging Technologies*, **9**, 348–354.

Koutchma, T., Keller, S., Chirtel, S. and Parisi, B. (2004) Ultraviolet disinfection of juice products in laminar and turbulent flow reactors. *Innovative Food Science and Emerging Technologies*, **5**, 179–189.

Kuo, F.L., Carey, J.B. and Ricke, S.C. (1997) UV irradiation of shell eggs: effect on populations of aerobes, molds, and inoculated *Salmonella typhimurium*. *Journal of Food Protection*, **60**, 639–643.

Kuse, D. (1982) UV-C sterilization of packaging materials in the dairy industry. *D. Milchwirtschaft*, **33**, 1134–1137.

Kwak, E.-J., Lee, Y.-S., Murata, M. and Homma, S. (2004) Effect of reaction pH on the photodegradation of model melanoidins. *LWT-Food Science and Technology*, **37**(2), 255–262.

Leeson, S., Díaz, G.J. and Summers J.D. (1995) *Poultry Metabolic Disorders and Mycotoxins*. University Books, Guelph, Ontario, Canada.

Liltved, H. and Landfald, B. (2000) Effects of high intensity light on ultraviolet-irradiated and non-irradiated fish pathogenic bacteria. *Water Research*, **34**, 481–486.

Lodi, R., Brasca, M., Mañaspina, P. and Nicosia, P. (1996) Improvement of the microbiological quality of goat milk by UV treatment. *Dairy Science Abstracts*, **58**, 484.

Lu, J.Y., Stevens, C., Yakabu, P., Loretan, P.A. and Eakin, D. (1987) Gamma electron beam and ultraviolet radiation on control of storage rots and quality of Walla Walla onions. *Journal of Food Processing*, **12**, 53–62.

Lu, G., Li, C., Liu, P., Cui, H., Yao, Y. and Zhang, Q. (2010) UV inactivation of microorganisms in beer by novel thin-film apparatus. *Food Control*, **21**, 1312–1317.

Luse, R.A. and McLaren, A.D. (1963) Mechanism of enzyme inactivation by ultraviolet light and the photochemistry of amino acids (at 2537 Å). *Photochemistry and Photobiology*, **2**, 343–360.

Lyon, S.A., Fletcher, D.L. and Berrang, M.E. (2007) Germicidal ultraviolet light to lower numbers of *Listeria monocytogenes* on broiler breast fillets. *Poultry Science*, **81**, 896–603.

Maharaj, R. (1995) The effect of ultraviolet radiation (UV-C) on the postharvest storage behaviour of tomato (*Lycopersicon esculentum* Mill cv. Capello). PhD Thesis, Laval University, Quebec, Canada.

Marquis, R.E. and Baldeck, J.D. (2007) Sporocidal interactions of ultraviolet irradiation and hydrogen peroxide related to aseptic technology. *Chemical Engineering and Processing*, **46**, 547–553.

Matak, K.E., Churey, J.J., Worobo, R.W., Sumner, S.S., Hovingh, E. and Hackney, C.R. (2005) Efficacy of UV light for the reduction of *Listeria monocytogenes* in goat's milk. *Journal of Food Protection*, **68**, 2212–2216.

Matak, K.E., Sumner, S.S., Duncan, S.E., Hovingh, E., Worobo, R.W., Hackney, C. R. and Pierson, M.D. (2007) Effects of ultraviolet irradiation on chemical and sensory properties of goat milk. *Journal of Dairy Science*, **90**, 3178–3186.

McLaren, A.D., Gentile, P., Kirk Jr, D.C. and Levin, N.A. (1953) Photochemistry of proteins. XVII. Inactivation of enzymes with ultraviolet light and photolysis of the peptide bond. *Journal of Polymer Science*, **10**(3), 333–344.

Mercier, J. and Arul, J. (1993) Induction of systematic disease resistance in carrot roots by pre-inoculation with storage pathogens. *Canadian Journal of Plant Pathology*, **15**, 281–283.

Micard, V., Belamri, R., Morel, M.H. and Guilbert, S. (2000) Properties of chemically and physically treated wheat gluten films. *Journal of Agriculture and Food Chemistry*, **48**, 2948–2953.

Milly, P.J., Toledo, R.T., Chen, J. and Kazem, B. (2007) Hydrodynamic cavitation to improve bulk fluid to surface mass transfer in a non immersed ultraviolet system for minimal processing of opaque and transparent fluid foods. *Journal of Food Science*, **72**(9), 407–413.

Morgan, R. (1989) UV green light disinfection. *Dairy Industries International*, **54**, 33–35.

Ngadi, M., Smith, J.P. and Cayoutte, B. (2003) Kinetics of ultraviolet light inactivation of *Escherichia coli* O157:H7 in liquid foods. *Journal of the Science of Food and Agriculture*, **83**, 1551–1555.

Nicolas, R. (1995) Aseptic filling of UHT dairy products in HDPE bottles. *Food Technology European*, **2**, 52–58.

Nigro, F., Ippolitto, A. and Lima, G. (1998) Use of UV-C light to reduce *Botrytis* storage rot of table grapes. *Post Harvest Biology and Technology*, **13**, 171–181.

Nkama, I., Mobbs, J.H. and Muller, H.G. (1987) Destruction of aflatoxin B1 in rice exposed to light. *Journal of Cereal Science*, **5**, 167–170.

Nkama, I. and Muller, H.G. (1988) Effects of initial moisture content and light intensity on the photo degradation of aflatoxin B1 in contaminated rice. *Journal of Cereal Science*, **8**(3), 269–274.

Neely, W.C. and West, A.D. (1972) Spectroanalytical parameters of fungal metabolites. 3. Ochratoxin A. *Journal of the Association of Analytical Chemistry*, **55**, 1305–1309.

Noci, F., Riener, J., Walkling-Ribeiro, M., Cronin, D.A., Morgan, D.J. and Lyng, J.G. (2008) Ultraviolet irradiation and pulsed electric fields (PEF) in a hurdle strategy for the preservation of fresh apple juice. *Journal of Food Engineering*, **85**, 141–146.

Onursal, C.E., Gozlekci, S., Erkan, M. and Yildirim, I. (2010) The effects of UV-C and hot water treatments on total phenolic compounds of juice, peel and seed extracts of pomegranates (*Puni granatum* L.). *Acta Horticulturae*, **877**, 1505–1509.

Oppenheimer, A.J., Jacangelo, J.G., Lane, J.M. and Hoagland, J.E. (1997) Testing the equivalency of ultraviolet light and chlorine for disinfection of wastewater to reclamation standards. *Water Environmental Research*, **69**, 14–24.

Ozen, B.F. and Floros, J.D. (2001) Effects of emerging food processing techniques on the packaging materials. *Trends in Food Science and Technology*, **12**, 60–67.

Panadés, R., Alonso, J.M. and Ibarz, A. (1997) Fotodescomposición de tiabendazol en solución acuosa. *Afinidad*, **470**, 299–304.

Pesek, C.A. and Warthesen, J.J. (1987) Photo degradation of carotenoids in a vegetable juice system. *Journal of Food Science*, **52**(3), 744–746.

Pesek, C.A. and Warthesen, J.J. (1990) Kinetic model for photo isomerization and concomitant photo degradation of b-carotenes. *Journal of Agriculture and Food Chemistry*, **38**, 1313–1315.

Quintero-Ramos, A., Churey, J.J., Hartman, P., Barnard, J., and Worobo, R.W. (2004) Modeling of *Escherichia coli* inactivation by UV irradiation and different pH values in apple cider. *Journal of Food Protection*, **67**, 1153–1156.

Rabek, J.F. (1982) *Experimental Methods in Photochemistry and Photophysics*. John Wiley & Sons, Inc., New York, USA.

Rhim, J.W., Gennadios, A., Fu, D., Weller C.L. and Hanna, M.A. (1999) Properties of ultraviolet irradiated protein films. *Lebensm.-Wiss. u.-Technol.*, **32**, 129–133.

Samarajeewa, U.Y. and Arseculeratne, S.N. (1974) Control of aflatoxin contamination-2. Detoxification of aflatoxin contaminated coconut oil. *Proceedings of the Ceylon Association Advances in Science*, **30**, 36.

Samarajeewa, U., Jayatilaka, C.L.V., Ranjithan, A., Gamage, T.V. and Arseculeratne, S.N. (1985) A pilot plant for detoxification of aflatoxin B1contaminated oil by solar irradiation. *Mircen Journal of Appl. Microbiology and Biotechnology*, **1**, 333–343.

Samarajeewa, U., Gamage, T.V. and Arseculeratne, S.N. (1987) Non-toxicity of solar-irradiated edible oils contaminated with aflatoxin B1. In: *Progress in Venom and Toxin Research*, Gopalakrshnakone, P. and Tan, C.K. (eds). University of Singapore, Singapore.

Samarajeewa, U., Sen, A.C., Cohen, M.D. and Wei, C.I. (1990) Detoxification of aflatoxins in foods and feeds by physical and chemicals methods. *Journal of Food Protection*, **53**(6), 489–501.

Seiji, M. and Iwashita, S. (1965) Enzyme inactivation by ultraviolet light and protective effect of melanin. *Journal of Biochemistry*, **57**(3), 457–459.

Shantha, T. and Sreenivasa Murthy, V. (1977) Photo-destruction of aflatoxin in groundnut oil. *Indian Journal of Technology*, **15**, 453–454.

Shantha, T. and Srrenivasa Murthy, V. (1980) Storage of groundnut oil detoxified by exposure to sunlight. *Indian Journal of Technology*, **18**, 346–347.

Shantha, T. and Sreenivasa Murthy, V. (1981) Use of sun light to partially detoxify groundnut (peanut) cake flour and casein contaminated with aflatoxin B1. *Journal of the Association of Analytical Chemists*, **64**, 291–293.

Shi, K., Kokini, J.L. and Huang, Q. (2009) Engineering zein films with controlled surface morphology and hydrophilicity. *Journal of Agriculture and Food Chemistry*, **57**, 2186–2192.

Sizer, C.E. and Balasubramaniam, V.M. (1999) New intervention processes for minimally processed juices. *Food Technology*, **53**, 64–67.

Snowball, M.R. and Hornsey, I.S. (1988) Purification of water supplies using ultra-violet light. In: *Development in Food Microbiology*, Robinson, R.K. (ed.). Elsevier Applied Science, London, pp. 171–192.

Sommer, R., Lhotsky, M., Haider, T. and Cabaj, A. (2000) UV inactivation liquid-holding recovery and photoreactivation of *Escherichia coli* O157 and other *Escherichia coli* pathogenic strains in water. *Journal of Food Production*, **63**, 1015–1020.

Stevens, C., Wilson, C.L., Lu, J.Y., Khan, V.A., Chalutz, E., Droby, S., Kabwe, M. K., Haung, Z., Adeyeye, O., Pusey, L.P., Wisniewski, M.E. and West, M. (1996) Plant hormesis induced by ultraviolet light-C for controlling postharvest diseases of tree fruits. *Crop Protection*, **15**, 129–134.

Stevens, C., Khan, V.A., Lu, J.Y., Wilson, C.L., Pusey, P.L., Igwegbe, E.C.K., Kabwe, M.K., Mafolo, Y., Liu, J., Chalutz, E. and Droby, S. (1997) Integration of ultraviolet a (UV-C)

light with yeast treatment for control of post harvest storage rots of fruits and vegetables. *Biological Control*, **10**, 98–103.

Stevens, C., Khan, V.A., Lu, J.Y., Chalutz, E.M., Droby, S., Kabwe, M.K., Huang, Z., Adeyeye, O., Pusey, P.L. and Tang, A.Y.A. (1999) Induced resistance of sweet potato to Fusarium root by UV-C hormesis. *Crop Protection*, **18**, 463–470.

Sumner, S.S., Wallner-Pendleton, E.A., Froning, G.W. and Stetson V.E. (1995) Inhibition of *Salmonella typhimurium* in agar medium and poultry skin by ultraviolet energy. *Journal of Food Protection*, **59**, 319–321.

Taghipour, F. (2004) Ultraviolet and ionizing radiation for microorganism inactivation. *Water Research*, **38**, 3940–3948.

Tran, T.T.M. and Farid, M.M. (2004) Ultraviolet treatment of orange juice. *Innovative Food Science and Emerging Technologies*, **5**, 495–502.

Unluturk, S., Atilgan, M.R., Baysal, A.H. and Tari, C. (2008) Use of UV-C radiation as a non-thermal process for liquid egg products (LEP). *Journal of Food Engineering*, **85**(4), 561–568.

Ustunol, Z. and Mert, B. (2004) Water solubility, mechanical barrier, and thermal properties of cross-linked whey protein isolate-based films. *Journal of Food Science*, **69**(3), FEP129–133.

Walking-Ribeiro, M., Noci, F., Cronin, D.A., Riener, J.G., Lyng, J.G. and Morgan, D.J. (2008) Reduction of *Staphylococcus aureus* and quality changes in apple juice processed by ultraviolet irradiation, pre-heating and pulsed electric fields. *Journal of Food Engineering*, **89**, 267–273.

Wallner-Pendleton, E.A., Sumner, S.S., Froning, G.W. and Stetson, L.E. (1994) The use of ultraviolet radiation to reduce psychrotrophic bacterial contaminations on poultry carcasses. *Poultry Science*, **73**(8), 1327–1333.

Whitby, G.B. and Palmateer, G. (1993) The effect of UV transmission. Suspended solids and photoreactivation on microorganisms in wastewater treated with UV light. *Water Science Technology*, **27**, 379–386.

Whited, L.J., Hammond, B.H., Chapman, K.W. and Boor, K.J. (2002) Vitamin A degradation and light-oxidized flavour defects in milk. *Journal of Dairy Science*, **85**, 351–354.

Wihodo, M. and Moraru, C.I. (2013) Physical and chemical methods used to enhance the structure and mechanical properties of protein films: a review. *Journal of Food Engineering*, **114**, 292–302.

Wolfe, R.L. (1990) Ultraviolet disinfection of potable water. *Environmental Science and Technology,* **24**, 768–773.

Worobo, R.W. (1999) Efficacy of the CiderSure 3500 ultraviolet light unit in apple cider. Department of Food Science and Technology Cornell University. In: *Apple Cider Food Safety Control Workshop*, US Food and Drug Administration, Centre for Food Safety and Applied Nutrition, July, pp. 15–16.

Wright, J.R., Sumne, S.S., Hackney, C.R., Pierson, M.D. and Zoecklein, B.W. (2000) Efficacy of ultraviolet light for reducing Escherichia coli 0157:H7 in unpasteurized apple cider. *Journal of Food Protection*, **63**(59), 563–567.

Wrong, E., Linton, R.H. and Gerrard, D.E. (1998) Reduction of *E. coli* and *S. seftenberg* on pork skin and pork muscle using ultraviolet light. *Food Microbiology*, **15**, 415–423.

Xu, P., Peccia, J., Fabian, P., Martyny, J.W., Fennelly, K.P., Hernandez, M. and Miller, S.L. (2003) Efficacy of ultraviolet germicidal irradiation of upper-room air in inactivating

airborne bacterial spores and mycobacteria in full-scale studies. *Atmospheric Environment*, **37**, 405–419.

Yousef, A.E. and Marth, E.H. (1985) Use of ultraviolet energy to degraded aflatoxin M1 in raw or heated milk with and without added peroxide. *Journal of Dairy Science*, **69**, 2243–2247.

Yousef, A.E. and Marth, E.H. (1987) Kinetics of interaction of aflatoxin M1 in aqueous solutions irradiated with ultraviolet energy. *Journal of Agriculture and Food Chemistry*, **35**, 785–789.

18

Application of Microwave Technology in Food Preservation and Processing

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18.1 Introduction

In this chapter, an overview is provided on the microwave preservation and processing applications in the food industry. The chapter explains the basics behind microwave heating, including food-related phenomena that influence microwave heating performance, with special concern on how food geometry and dielectric properties of foods affect the microwave heating characteristics. It also gives examples of microwave preservation and processing applications in the food industry, as well as a section on the bottlenecks and future trends. This chapter provides a starting point to the subject; the interested reader can find more information on details in the references.

18.2 Background

Applications for microwave processing of foods have been considered highly promising and novel for well over 40 years. The number of operating microwave lines in the food industry is probably more than one thousand

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worldwide (Bengtsson, 2001) and has been growing steadily during the latest decade. The technology has also been introduced industrially for a wider range of applications during this time period. Many applications have been claimed successful on a laboratory and pilot scale over the years, with advantages of greatly reduced processing times and much improved yield and product quality.

However, until recently, there was a relatively slow growth in the number of industrial installations. Among the reasons for this slow growth are many factors:

- Earlier, little consideration was given to what reference method was being used, and whether this was the optimal method or could easily be improved upon, eliminating much of the advantage of the microwave alternative.
- The microwave equipment manufacturers often had no experience in designing processing equipment for the food industry's specific needs. It is possible that personnel in the food industry lacked knowledge on microwave heating fundamentals, and is thus sceptical to the new technology.
- Earlier, solutions for scaling-up were more difficult. Today, however, powerful simulation tools make it possible to find solutions for scaling-up in a more straightforward way.

There are many requirements for a viable industrial microwave heating application:

- a high degree of reliability,
- advantages based on some of the unique features of microwave heating that cannot be easily matched by an improved conventional technique,
- cost-effectiveness in terms of high power efficiency,
- application development and scale-up performed in close cooperation between microwave equipment manufacturers, specialists on process equipment and control systems for the food industry, and the technical staff of the food company.

The large number of installations for microwave tempering, drying and pasteurization is now being followed by applications on microwave baking of crust-less bread, sterilization of ready meals and cooking of desserts, due to successful cooperation between food companies, food technologists, microwave engineers and equipment suppliers. Several good examples of successful industrial implementations of microwave preservation and processing exist. This will be further described in Section 18.4.

18.3 Principles

18.3.1 Electromagnetic basics of microwave heating

Electromagnetic waves Electromagnetic waves with frequencies in the range from 300 MHz to 300 GHz are defined as microwaves, with the corresponding wavelengths between 1 m and 1 mm, respectively. Microwaves used in the food industry for heating are the industrial, scientific and medical (ISM) frequencies of 2450 MHz or 915 MHz,[1] corresponding to a 12 or 33 cm wavelength in vacuum. Most foods contain a substantial amount of water. The molecular structure of water consists of two slightly positively charged hydrogen atoms and a negatively charged oxygen atom, which altogether form a dipole. When a microwave electric field is applied to a food, dipoles, mainly water, and ionic components (the latter caused mainly by dissolved salts in the food) try to orient themselves to the electric field (similar to a compass in a magnetic field). Since the rapidly oscillating electric field changes from positive to negative and back again, in several million times/second, the dipoles try to follow and these rapid movements correspond to heat generation. The increase in temperature of water molecules gives rise to heat in surrounding components of the food by conduction and/or convection. Because of their widespread domestic use, some popular notions have arisen that the microwaves 'heat from the inside out'. What actually occurs is that outer parts receive the same energy as inner parts, but the surface will lose its heat faster to the surroundings by evaporative cooling. The distribution of water and salt within a food will have the major effect on the amount of heating, although differences also occur in the rate of heating as a result of the shape of the food.

The penetration depth d_p of the microwave energy is determined by the complex permittivity $\epsilon_r = \epsilon_r' - j\epsilon_r''$ (often referred to altogether as dielectric properties)[2]:

$$d_p = \frac{1}{2\pi f \sqrt{2\mu_0\epsilon_0\epsilon' \left\{ \sqrt{1 + \left(\frac{\epsilon''}{\epsilon'}\right)^2} - 1 \right\}}} \tag{18.1}$$

where f is the microwave frequency (Hz), μ_0 is the permeability in vacuum ($4\pi \times 10^{-7}$ V s/A m) $\approx 1.257 \times 10^{-6}$ H/m) and ϵ_0 is the permittivity in vacuum

[1] In Europe (except in the UK) and in several of the Pacific rim countries, there is no allocated ISM frequency band at 915 or 896 MHz. In the UK, 896 MHz is used instead of 915 MHz. In the USA and Canada, 915 MHz is used (also with limited use in Australia and New Zealand).

[2] It should be noted that this definition is based on the assumption of normal incidence of a plane wave at an infinitely plane material; the penetration depth therefore only gives an indication of the heating characteristics; in a practical situation, other phenomena like resonance phenomena which are strongly correlated to e.g., geometrical conditions, size of the food etc., must also be taken into account.

(8.854×10^{-12} F/m). The conductivity σ (S/m) is related to the effective loss factor ϵ''_{eff} according to

$$\sigma/(\omega\epsilon) = \epsilon''_{eff}/\epsilon'_r \tag{18.2}$$

These properties have been recorded for some foods (Kent, 1987; Ohlsson and Bengtsson, 1975; Bengtsson and Risman, 1971). They vary with the moisture content and temperature of the food and the frequency of the electric field. In general, the lower the loss factor (that is greater transparency to microwaves) and the lower the frequency, the greater is the penetration depth. The penetration ability of microwaves in foods is in some cases limited. For high-moisture foods, the penetration depth is approximately 1–2 cm at 2450 MHz. At higher temperatures, the electric resistance heating due to dissolved ions will also contribute to the heating mechanisms, normally by further reducing the penetration depth of the microwave energy. The distribution of energy and heat within the food will depend on several interacting factors: geometrical and size conditions of the food, placement of individual food components in relation to each other, the dielectric properties (complex permittivity) of the food, as well as the microwave frequency (see the next section). In order to have an idea of how a specific food will be heated by microwaves in a microwave oven or industrial equipment, simulations of the microwave heating distribution are therefore often used. The control of the heating uniformity of microwave heating is complicated (Wäppling Raaholt, Risman and Ohlsson, 2006, 2001) and of major concern to achieve successful industrial implementations of microwave applications in the food industry. However, modelling tools have made a breakthrough as industrial microwave equipment can now be designed based on modelling results, so as to provide as uniform microwave heating distribution as possible.

Fields in a cavity bounded by metal: influence of microwave oven

Field patterns in single-mode and multimode cavities During microwave heating of foods, the food items are enclosed in spaces or cavities surrounded by metal walls. Those specially designed cavities are commonly referred to as microwave cavities. A microwave cavity can be categorized as single-mode or multimode. A single-mode cavity has one dominant mode and is a result of the cavity dimensions and the feeding of the microwave energy. The single-mode cavity is the primary cause for the heating field pattern. This field pattern is created by the standing wave (Hecht, 1989; Cheng, 1989) between the walls of the cavity. Typically, the size of the cavity is comparable to, or slightly larger than, that of the waveguide, and the excitation frequency from the microwave source is provided within a narrow frequency band to maintain the necessary coupling (Metaxas and Meredith, 1983). A single-mode cavity, as the one described by Schubert and Regier (2005), may have the advantage of giving a maximum electric field in the centre of the cavity, while on the other hand a

disadvantage being that the zone in which the food material can be effectively heated is relatively small. This design could be used for microwave heating of small samples in analytical laboratories or for heating liquid or semi-liquid pumpable materials in industrial applications (Isaksson, 2002, Ohlsson, 1993). Multimode cavities, on the other hand, are most commonly used in microwave heating applications, with a typical domestic microwave oven as an example. In general, the dimensions of a multimode cavity are several times larger than the free space wavelength of the microwave field generated by the magnetron. In a multimode cavity, several different field patterns are possible over a narrow frequency band (Balanis, 1989), where each field pattern represents a given microwave mode. The cut-off frequency is the lowest frequency that allows propagation of an electromagnetic wave (Cheng, 1989). In a cavity, the cut-off frequency is different from that in a waveguide, since a waveguide is open-ended while a microwave cavity is enclosed. The cut-off frequency ($f_{c,mnp}$) for a microwave cavity is calculated from

$$f_{c,mnp} = \frac{1}{2\pi\sqrt{\mu\sigma}}\sqrt{\left\{\left(\frac{m\pi}{a}\right)^2 + \left(\frac{n\pi}{b}\right)^2 + \left(\frac{p\pi}{c}\right)^2\right\}} \tag{18.3}$$

where m, n and p are integers that describe the discrete pattern of the half-wave variation of the microwave field, with corresponding lengths a, b and c along the x, y and z axes, respectively (Metaxas and Meredith, 1983). The microwave modes in an empty microwave cavity are referred to by the discrete pattern of m, n and p, representing the x, y and z directions. These modes are designated as TE_{mnp} and TM_{mnp} for transverse electric (TE) and transverse magnetic (TM), respectively. Several TE and TM modes may exist at the same time at a specific frequency bounded by its corresponding cut-off frequencies. These modes are defined as degenerate modes. However, two different modes (TE and TM) will only exist at the same frequency if their indices (m, n and p) are nonzero or if two cavity sides are equal in length. Although different modes may exist for the same frequency, their corresponding field distribution will not be the same. The possible modes that may exist in a microwave cavity can be estimated by using

$$2\pi f_{mnp}\sqrt{\mu\sigma} = \sqrt{\left\{\left(\frac{m\pi}{a}\right)^2 + \left(\frac{n\pi}{b}\right)^2 + \left(\frac{p\pi}{c}\right)^2\right\}} \tag{18.4}$$

By applying this to an empty microwave cavity, with cubical shape ($a = b = c$), this equation simplifies to

$$4\left(\frac{a}{\lambda}\right)^2 = m^2 + n^2 + p^2 \tag{18.5}$$

The left-hand side of Equation (18.4), with the operating frequency range f_{mnp} and the dimensions of the microwave oven (a, b, c) inserted gives the possible microwave modes in the cavity and their corresponding indices m, n and p. A combination of m, n and p which gives a value within the range of the left-hand side of Equation (18.4) is a valid index.

Applicators A microwave applicator is a device that is used to couple the microwave energy into the material to be heated. Applicators may have different configurations and could be designed based on the properties, the shape/size and the volume of the material to be heated. The three most common applicator types are:

- *Resonant applicators* are the most commonly used form of microwave heating applicators and are classified as either single-mode or multimode. A single-mode applicator develops only one basic microwave field pattern inside the cavity (Cheng, 1989; Balanis, 1989), which focuses the heating to a well-defined area. The product (or load) must be placed in, or moved to, this area. In multimode applicators, multiple field patterns are generated inside the cavity (Metaxas and Meredith, 1983), which allows for a more uniform but less well-defined heating of larger materials. An example of a multimode cavity is a household microwave oven. Microwave energy is introduced into a closed metal box, a cavity. In order for the heating uniformity to be improved further, a small rotating propeller (called a mode stirrer) can be used, or a rotating plate where the product is placed.
- *Travelling wave applicators* are typically constructed from rectangular or cylindrical waveguides and match the source to the load. The material or load is heated by the microwave energy present inside the waveguide. Any microwave energy that does not couple to the load is absorbed by an attached load at the end of the waveguide.
- *Near field applicators* are classified by an open-ended waveguide, or a waveguide with slots, which allows passage of the microwave energy. The small distance between the applicator and the load gives rise to their classification as being 'near field'. Typical applications utilizing near field applicators include heating of materials on a conveyor belt.

Applicator design for food application is complex and needs to be done taking into account the interaction between parameters important for heating uniformly, such as food composition and geometry, packaging geometry and composition, and cavity dimensions.

Fields in lossy dielectrics A lossy dielectric medium is defined as a medium in which the electrical conductivity is not equal to zero; yet the medium is not a good conductor. According to the definition of complex permittivity,

the dielectric loss of such a medium is significantly higher than for a good conductor.

Microwave heating of foods has sometimes been associated with uneven heating, due to the so-called 'hot and cold spots', which may be present in the food product after heating. The microwave heating profile in foods is determined by the thermophysical properties of the food item (the dielectric properties and the thermal properties) as well as of the distribution of the absorbed microwave power in the food. The latter is, in turn, determined by several factors: the electric field inside the microwave cavity or applicator, the dielectric properties of the food item, but also by the microwave frequency. Computational modelling-based design of the oven cavity (e.g. size and shape of the cavity or applicator), as well as the waveguide system, gives tremendous possibilities to control the electromagnetic field pattern. The resulting heating uniformity depends on several interacting parameters such as food composition, size and geometry, packaging geometry and composition, and cavity dimensions. This will be described further in the next subsection. In the literature, more on the general principles for microwave heating can be found. Several authors have described the subject, for example Wäppling Raaholt and Ohlsson (2009, 2005), Ohlsson and Bengtsson (2001), Buffler (1993), Walker (1987), Ohlsson (1983), Ohlsson and Risman (1978), and Bengtsson (1971).

Influence of food geometry and dielectric properties on heating performance

Factors influencing the microwave heating uniformity Several interacting variables related to food, package and the microwave oven itself will influence how the food will be heated. Particularly multi-component foods often require tailor-made product development, aiming at avoiding tendencies to heat unevenly, which might otherwise cause problems with both sensory and microbiological quality.

In this section, an overview is provided on food-related phenomena that influence microwave heating performance, with special concern to how food geometry and dielectric properties of foods are affecting the microwave heating characteristics. Both geometry and size of different components, as well as recipe formulation and relative placement of different components could be regarded as food parameters. This section provides an introduction to these matters; the interested reader can find more information on the details in the references (e.g. Wäppling Raaholt and Ohlsson, 2009).

Dielectric properties The knowledge of the dielectric properties of the foods to be microwave processed is essential for an appropriate design of microwave applicators and equipment. The property that describes the behaviour of a dielectric under the influence of a microwave field is the complex permittivity:

$$\epsilon^* = \epsilon' - j\epsilon''_{eff} \tag{18.6}$$

whereas the absolute permittivity is given by

$$\epsilon = \epsilon_0 \epsilon^* \tag{18.7}$$

where ϵ' is the dielectric constant and ϵ''_{eff}, is the effective loss factor. When microwave heating of foods occurs, both dipole redistribution and ionic conduction contribute to the heating mechanisms (Mudgett, 1995; Metaxas and Meredith, 1983; Mudgett *et al.*, 1980; Nelson, 1973). The so-called loss mechanism, which corresponds to heating, is characterized by the relative loss factor term ϵ''_{eff}, which is part of the complex relative permittivity. The complex permittivity is often called the dielectric properties.

For food loads heated at 2450 MHz, both loss mechanisms due to ionic conduction (like in salty foods) as well as dielectric relaxation are present. The effective loss factor $\epsilon''_{effective}$, where both types of losses are included, is then often used (Metaxas, 1996).[3] The following relationship then describes the relative effective loss factor:

$$\epsilon''_{effective} = \frac{\sigma}{\omega \epsilon_0} + \epsilon'' \tag{18.8}$$

where σ is the electrical conductivity in S/m, ω is the angular frequency in radians/second, ϵ_0 is the absolute permittivity of free space and ϵ'' is the loss factor. The time-average value of the dissipated power density p_{diss} (W/m^3) in typical food products is proportional to the square of the electric field:

$$p_{diss} = \frac{1}{2} \mathrm{Re} \left\{ \vec{E} \cdot \vec{J}^* \right\} = \frac{1}{2} \omega \epsilon_0 \epsilon''_{effective} \left| \vec{E} \right|^2 \tag{18.9}$$

where $\vec{J}^*$ is the complex conjugate of the electric current density (A/m^2), $\epsilon''_{effective}$ is the relative effective loss factor of the food material and $\left|\vec{E}\right|$ is the amplitude (peak value) of the electric field intensity (V/m). An assumption here is that the relative complex permeability μ of the food material is assumed to be 1, that is the food is magnetically close to vacuum. In other words, we assume that foods do not contain metal.

The thermal properties of the foods will also contribute to the final heating distribution, by levelling out the temperatures between regions that have been heated more than their surroundings. More on thermal properties is found in Incropera and Dewitt (2002).

There are several methods to measure dielectric properties: cavity perturbation methods (Ohlsson and Bengtsson, 1975; Ohlsson *et al.*, 1974; Bengtsson and Risman, 1971; Risman and Bengtsson, 1971), open-ended

[3] For applications related to microwave heating of foods, the loss factor refers to two types of losses into heat: the dipole relaxation and the ionic conductivity. At other frequencies, however, other mechanisms may be more dominant (Hasted, 1973).

coaxial methods (Gabriel, Chan and Grant, 1994; Gabriel, Grant and Young, 1986), retro-modelling (Wäppling Raaholt and Risman, 2003; Risman, 1999) and shorted waveguide methods (also called von Hipple methods; von Hipple, 1954). Waveguide methods belong to the transmission line methods, where a sample is shaped to completely fill the cross-section of a transmission line (a coaxial or a rectangular waveguide). More on this may be found in Goedeken, Tong and Virtanen (1997). The choice of method may depend on several factors; these are the considered range of permittivity data and frequency, the required accuracy, the temperature range of interest, whether the food is nonhomogeneous or homogeneous, whether taking samples of the food (e.g. in a specific geometry) is easy or complicated and whether the food expands or contracts during the temperature interval of interest (e.g. during freezing or drying, respectively). Furthermore, the evaluation method for transferring the measurement data into dielectric data may also be a contributing factor.

Heating phenomena that influence heating performance and uniformity

In-depth heating Sometimes the academic idea of an infinite food, distributed in two directions, is used. With an incident wave on such a slab, the power level will gradually decrease inwards to an insignificant level if the slab thickness is large in comparison to the calculated penetration depth. For simplicity it may be assumed that the microwave field is evenly distributed and of a given field strength at the food surface. For a thinner slab, the remaining power level from microwaves that are impinging on the two opposite surfaces will overlap. This may result in more rapid heating of the central parts than of the surface regions. This takes into consideration that the surface will be cooled both by heat transfer to the surrounding air space and by evaporative cooling. Furthermore, internal standing waves may occur within a food slab by reflection from the back surface of the slab.

For layered materials of different dielectric properties, the microwaves will be reflected and refracted also at the interface between these materials, for example between meat and an outer layer of fat. Depending on the thickness and dielectric properties of the layers, standing wave patterns may be developed; hence, the fat layer will be overheated, in spite of its lower dielectric loss factor. A contributing factor will then be the much lower specific heat of the fat material.

For inhomogeneous food (based on composition) or if different food materials are heated side by side, temperature differences will result from the combined differences in dielectric and thermal properties although uniform microwave field strength may be anticipated.

Concentration effects A food item with sharp corners and edges that is microwave heated will show field and energy concentrations there, which cause selective heating, especially at the corners. Briefly described, a sharp

edge or corner will act as an antenna and attract more energy than surrounding areas. Considering microwave heating as electromagnetic waves, which may be reflected, absorbed or refracted, part of the energy is then reflected at the food surface, part of the energy is refracted while part of the refracted energy will be absorbed, as understood from elementary electromagnetics (Cheng, 1989). Most of the remaining energy will be reflected back to other food surfaces. Depending on the food geometry, the result can be on focusing energy to certain areas, which may be part of the explanation for so-called *concentration heating effects*.

For cylindrical or spherical geometries, such concentration effects can cause concentration of energy to the centre of the food, depending, however, on the food diameter and the dielectric properties. This centre overheating effect occurs for diameters of approximately 1 to 3 times the penetration depth in the material. For cylinders, concentration effects occur when the electrical field is parallel to the cylinder axis. The effect will be stronger for foods with high permittivity values. At 2450 MHz, centre heating usually happens at diameters of 25 to 55 mm, while the values are correspondingly larger (about 2.5 times larger) for 915 MHz.

Microwave heating performance is thus affected by several possible heating phenomena. Among these are *edge overheating*, *run-away heating* in frozen foods and *standing wave patterns* in food loads. Edge overheating is a result from electric fields of strong amplitude, while field vectors are parallel to an edge of a food item.

Run-away heating Pure ice has much lower dielectric properties than thawed foods. Most of the water in frozen foods is found as ice crystals within the food item. Since the regions that are thawed in the very beginning of a microwave thawing process will have much higher loss factors ϵ'', as compared to still frozen regions, they will heat very rapidly. This phenomenon is called thermal run-away heating (Buffler, 1993). As the product temperature is increased, from the frozen state to around −2 °C (for several types of foods of normal salt content), the dielectric constant and loss factor increase rapidly. Above freezing, both the loss factor and the dielectric constant decrease with increasing temperature. As the salt concentration increases, ionic concentrations may compensate for this decrease. For high-salt foods, such as salted ham, the dielectric constant will increase with temperature from above the freezing point to approximately 60 °C. Run-away heating could occur also in high-salt foods (Bengtsson and Risman, 1971).

Edge and corner overheating The so-called edge (or corner) overheating effect is often noted, especially when frozen foods are heated or thawed. Edges and corners have a tendency to heat or thaw first, which intuitively could be understood by the fact that the amplitudes of the electromagnetic fields are concentrated at such areas of the food, due to scattering phenomena (Figure 18.1 and

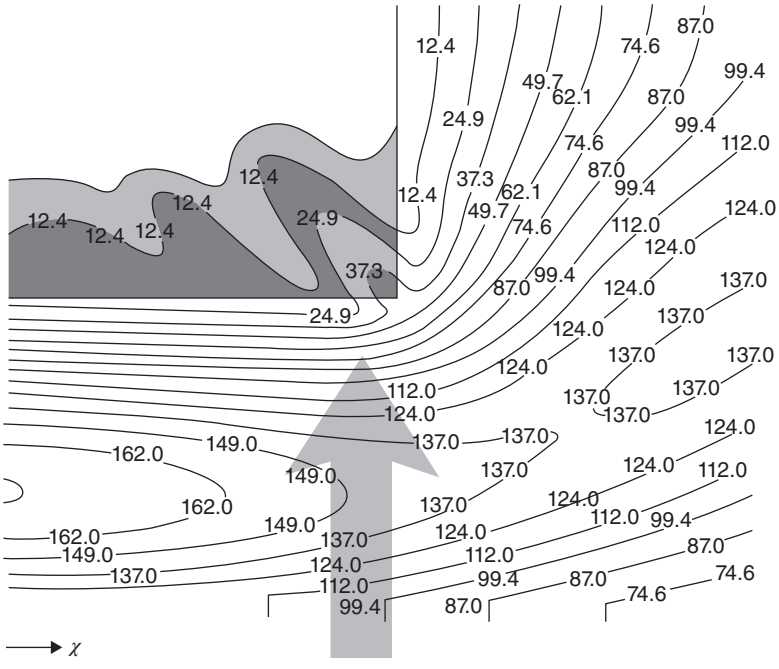


Figure 18.1 Illustration of the phenomenon behind edge overheating. The electromagnetic fields are concentrated at the corners of the food, due to scattering phenomena (source: Sundberg 1998. Reproduced with permission of SIK, Sweden)

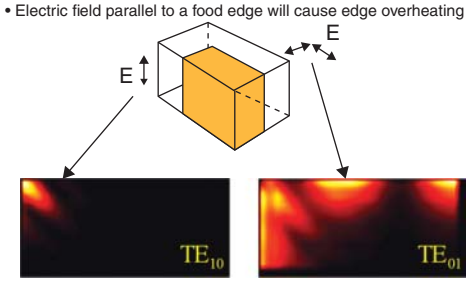


Figure 18.2 Illustration of edge overheating, where the electric field parallel to a food edge will cause edge overheating (source: Sundberg 1998. Reproduced with permission of SIK, Sweden) *See plate section for colour version*

18.2). At the boundary between the food load and the surrounding air, one of the boundary conditions in the solution of the electromagnetic problem leads to the continuity condition of the parallel component of the electric field. Edge overheating is strongly influenced by the polarization and incident angle of the incident field, the angle and curvature of the edges, the permittivity of the heated food materials and the presence of other scatterers close to the edge (Sundberg, 1998).

At the food edges, microwaves approach the food from two directions. Furthermore, parallel to a surface at the boundary, the electric fields have two polarizations. The resulting concentration of the energy distribution to the sharp edges is explained by the continuity condition at the boundaries of the parallel electric fields. This heating phenomenon is one of the dominating ones, related to heating uniformity problems in rectangularly shaped foods. For corners, the corresponding condition will analogously be the case for electric fields of three polarizations. This results in even more pronounced heating at the corners. By optimizing the food product design by means of modelling tools (Wäppling Raaholt, 2000; Wäppling Raaholt and Ohlsson, 2000), it is often possible to avoid corner and edge overheating. In several cases, more than 15% of the microwave energy that is absorbed by the food item is lost by the edge overheating phenomenon (Risman, 1992). The edge overheating effect is investigated for high-permittivity dielectrics in the work by Sundberg (1998).

Centre overheating For foods that are spherically or cylindrically shaped, that is foods with convex surfaces, refraction and reflection phenomena will result in concentration of the microwave power distribution to the geometrical centre for certain diameters (Ohlsson and Risman, 1978). This phenomenon is called centre overheating. It is influenced by different factors, mainly the geometries, sizes and complex permittivity values of foods. The phenomenon of centre overheating is exemplified in Wäppling Raaholt and Ohlsson (2005) and Ohlsson and Risman (1978).

Standing wave patterns in microwave heated foods Several different types of standing wave phenomena are occurring in foods during microwave heating (Ryynänen, Risman and Ohlsson, 2004). Standing waves may appear for the following cases:

- *between* the plane upper and lower surfaces,
- *within* certain thick loads (this latter phenomenon will be due to internal resonances, i.e. standing waves),[4]
- *at* larger surfaces and, finally,
- *at* edges of larger surfaces (which in turn is partly a result of the edge overheating effect and partly of surface wave phenomena).

The internal 'hot and cold spots' in the food item, that is the standing wave patterns of absorbed and reflected electromagnetic waves, can be quantified by modelling for simplified scenarios, as described by Ryynänen, Risman and Ohlsson (2004). This was performed by using an extension of the transverse

[4] It should be noted that external resonance phenomena, like the exploding egg effect, is not a standing wave phenomenon.

resonance method (Harrington, 1961) in order to include the behaviour of all TE and TM waveguide modes propagating in the vertical direction. Other kinds of phenomena are also occurring during microwave heating of foods. Examples of such phenomena are those that are related to different penetration depths for different kinds of food materials. This is described for multi-component foods in the following subsection.

Effects of geometry and dielectric properties It may be assumed that microwave ovens are designed to give as even a field distribution as possible in the loaded oven and to support TM microwave modes around the food sample in order to minimize corner and edge overheating. Then food composition, geometry and positioning as well as packaging will constitute the main remaining significant factors that determine heating performance and resulting food quality.

In general, for slab-shaped foods, product thickness should be even, and preferably limited to less than 2.5 times its microwave penetration depth at the specified frequency. Rounded edges and corners will limit preferential tendencies to corner and edge overheating. The hot area is usually found diagonally from the edge or corner some 7–8 mm into the food (Ohlsson, 1993). So-called 'hot and cold spots' may also be present. These are shown as temperature variations over large, flat surfaces of food and are normally larger than the in-depth temperature variation. Therefore, such 'spots' may often have a larger influence on the overall heating results. When heating food components of different dielectric and thermal properties such as in ready-to-heat meals on a tray or compartmented plate, the geometry and size, as well as the relative arrangement of the components, must be selected in order to optimize the heating uniformity. Advantages may often be taken of the tendencies to focus the electromagnetic fields to the centres of rounded (cylindrical or semi-spherical) foods. This is particularly true for thick food samples of limited penetration depth. To even out temperature distribution, food components with a high dielectric loss, for example meat stew, may be partially shielded by food components of lower loss (e.g. mashed potato). Material with a low dielectric loss is also less susceptible, since low ϵ'' will reduce the consequences of field concentrations due to the resulting slower temperature rise. The formation of variable standing wave patterns inside the food can be kept to a minimum by adjusting the thickness of the material with a low dielectric loss (Ryynänen and Ohlsson, 1996; Ohlsson and Thorsell, 1985).

In layered materials, standing wave patterns may develop as a result of microwave reflection at the component interfaces (Wäppling Raaholt and Ohlsson, 2005). In terms of 'controlling' the energy distribution and heating effects, advantage could be taken by the choice of layer materials and thickness of the layers. Ryynänen, Risman and Ohlsson (2004) described the situation when a hamburger is heated, sandwiched between two pieces of bun. A standing wave pattern will develop between the microwave oven top and the bottom walls, but also between the oven shelf and the bread

as well as the meat. However, since the food or dish must have a 'natural' appearance for consumer acceptability, the extent to which food geometry and positioning can be modified is sometimes fairly limited.

Furthermore, for multicomponent ready meals, the difference in dielectric and thermophysical properties of the components will influence the heating result**,** as will microwave reflections due to the boundaries between different components (Mudgett, 1986). The latter phenomenon is caused by the fact that each component will differ in microwave penetration depth (d_p) due to the different relative permittivities between the components. The microwave reflections at the interfaces between the air and food material will depend on the permittivity for each component, frequency, as well as on the microwave oven mode properties. Several different *selective heating phenomena*, like edge overheating, centre overheating and run-away heating, will also play a role. These types of phenomena have been further described in the previous subsections.

Surface browning and crisping Another means of influencing heating performance is by modifying the food composition or formulation, which in turn will change the dielectric properties. Microwave absorption will tend to be reduced and the microwave penetration depth in the food will be raised by lowering the water content and/or mixing with material of low permittivity. Increasing the ionic content, such as by adding salt, will increase dielectric loss and reduce the penetration depth without affecting the wavelength in the food. However, the sensory demands for natural taste, flavour and texture will put rather narrow limits on what changes in dielectric or thermal properties can be achieved.

To compensate for lacking flavour development, the addition of flavour substances (natural flavours, spices, etc.) may be required; for example, microencapsulated flavours are recommended with controlled release above a certain temperature during re-heating. For foods in which warm swelling starches are being used, the temperature reached during microwave heating may not be sufficient everywhere for such starches to swell, resulting in a raw taste and undesirable texture. As noted by Katt (1991), a combination of warm swelling and cold swelling modified starches is sometimes presented as the answer to this problem.

Microwave heating distribution and uniformity There are modelling-based methods available (Wäppling Raaholt, 2000) for designing microwave foods into products that give a more uniform heating distribution. By appropriately combining suitable food parameters (placement, geometry and amount of food components) in microwave ready meals, the heating uniformity could be considerably improved. The starting point of the method is a defined measure of heating uniformity in the food, which is then used when modelling the electromagnetic fields to predict and control the heating uniformity. As a result, ready-to-heat microwave meals with a resulting more levelled-out

temperature distribution could be designed and in a next step produced in the food industries.

Modelling and simulation of the electromagnetic fields to predict the heat distribution in the foods involves the solution of Maxwell's equations (Maxwell, 1954) in three dimensions. Today, several different modelling regimes exist, including the FDTD (finite-difference time-domain), the FETD (finite-element time-domain) and the FVTD (finite-volume time-domain) modelling. Furthermore, frequency domain modelling is also quite common, even if for modelling of microwave heating the time-domain is probably the most widespread modelling technique.

Practical validation of the modelling results is an important step towards development of uniformly microwave heated foods. For this purpose, the on-line temperature during microwave heating may be performed by fibre-optic probes by employing surface measurement methods in terms of thermography or by liquid crystal foils. Furthermore, alternative indirect techniques are also available, for example modelling foods that shifts colour at a predetermined temperature (Wäppling Raaholt, 2009; Risman, Ohlsson and Lingnert, 1993; Ohlsson, 1981). In addition, MRI has the potential to give a useful heating pattern (Nott *et al.*, 1999). Improved modelling techniques and instrumentation will altogether give an additional increase in the number of industrially implemented microwave food applications.

Appropriate equipment and process design is necessary to guarantee that the technologies are adapted to the product and food application in question. A successful development of tailor-made and reliable equipment for microwave processing requires an effective collaboration between microwave technology specialists, equipment suppliers and food technologists.

18.4 Applications of microwave in food preservation and processing

Microwave heating was rapidly adopted by the food industry in the 1950s, after the issue of the first patent, which described an industrial conveyor belt microwave system in 1952. The main successful applications of microwave technology in the food industries are shown in Table 18.1, while Table 18.2 shows the advantages and disadvantages of microwave heating.

A successful implementation of microwave technology can bring many benefits to the food producer; however, the success of the implementation is set in the early phases of process development. Over the years, many industrial applications have been developed and abandoned due to processes that do not meet the needs and specifications of the food processing plant. As all technologies, microwave processing has its specific advantages and limitations. Due to the fact that the underlying physical phenomena of microwaves are not known to the typical food processor, and that the food manufacturing requirements are not always understood by the microwave equipment designer and supplier,

Table 18.1 Successful applications of microwave technology in the food industries (see Schiffman, 2001; Bengtsson, 2001; Decareau, 1986; Metaxas and Meredith, 1983)

Process	Products
Tempering-thawing	Meat, poultry, berries, fruits, butter, fish
Melting and rendering	Chocolate, fat, tallow, lard, residues from meat industry
Blanching	Vegetables (e.g. corn), potatoes, fruit
Drying	Pasta, onions, rice cakes, juice, potato chips, egg yolk, snack foods, seaweed
Vacuum and freeze drying	Beverages, fruit juice (e.g. orange juice), grains, seeds, heat-sensitive products, powders, meat, vegetables, fruits, carrots
Puffing	Starch-based products (e.g. popcorn)
Baking	Proofing of bread dough, baking of bread and cakes, final baking of cookies and rusks, doughnut proofing and frying, potatoes
Roasting	Coffee beans, peanuts and other nuts, cocoa beans
Disinfection (insects and microbes)	Cereals, packaging material
Cooking	Meat, fillets, sausages, meat pie, sausage, potatoes, chicken, bacon, sardine, herring, potato crisps, rice
Pasteurization	Bread, yoghurt, ready-made meals, peeled potatoes, crab-fish, beverage, milk
Sterilization	Yoghurt, pre-cooked foods, bread, pizza, peeled potatoes, grain, coconut flesh

Table 18.2 Advantages and disadvantages of microwave food processing

Advantages	Disadvantages
• Very fast in-depth heating • Significantly shorter process time • Energy effective process due to shorter process time and effective product heating • Suitable for batch or continuous processing • Improved product quality for several types of applications • Clean technology	• Inhomogeneous food heating/focusing, e.g. for cases where food products are not properly designed for its purpose • Special security measures are necessary in the equipment to avoid leakage • Special protection of electronic parts for heating and humidity • Relatively high equipment cost, which, however, should be put into the context of the often rather short payback time of an investment • Metals in packaging should often be avoided

it is of great importance to identify the needs and technical specifications that have to be met and to involve external know-how and resources necessary at early stages of the developmental process. A generalized process development flowchart highlights some of the important considerations when developing industrial microwave applications (Holtz and Raaholt, 2013).

18.4.1 Pasteurization and sterilization

Heating uniformity is important for all microwave systems, which requires appropriate design of the system, including a waveguide system, applicators and cavities. For pasteurization and sterilization, uniformity in heat is, however, crucial to ensure that microbiological requirements are fulfilled. Modelling tools can be used to predict the location of 'cold spots'. However, validation by measurements is always needed to confirm sufficient heat treatment.

Since microwave energy can be used effectively and rapidly to heat foods that contain water or dissolved salts, the application of microwave pasteurization or sterilization has been frequently studied. The major advantages of microwave heating are rapid heating, time savings and volumetric heating. The high and homogeneous heating rates in some foods and the corresponding short process times can offer a high food product quality. Microwave applications for pasteurization or sterilization include packed foods (e.g. ready meals), heated continuously on a conveyor belt or batch-wise, while continuous preservation of fluid or semi-fluid pumpable foods is also possible, and offers an alternative to conventional systems like a scraped surface heat exchanger.

Pasteurization and sterilization of ready meals and packed foods Conveyor belt systems for microwave pasteurization of ready meals are industrially implemented worldwide. An example of a microwave pasteurization line for continuous heat treatment of ready meals is found in Figure 18.3 (www.micvac.com).

An HTST (high temperature–short time) process should result in quality advantages with maintained safety, both in the pasteurization and sterilization of packaged foods (Ohlsson and Bengtsson, 2001), as demonstrated

Figure 18.3 Microwave pasteurization tunnel for production of ready-to-heat meals (source: http://www.micvac.com/food-technology/equipment by Hanna Rüdel. Reproduced with permission of MicVac.)

by the relationships established between time, temperature, inactivation of microorganisms, enzymes and sensory and nutritional losses. Microwave heating of packaged foods is a heating method by which HTST-like processing can be achieved for solid foods of several centimetre thickness. Important considerations to such a microwave process are what minimum temperature spread can be controlled and the fact that the cooling rate will depend on conventional cooling techniques and the thermal conductivity of the food material. For such installations, a maximum temperature spread of 5 °C has been claimed (Ohlsson and Bengtsson, 2001).

In a previous EU project (EC, 2000), where SIK, Campden and Chorleywood RA, the Fraunhofer Institute and industrial partners participated, the benefits of microwave sterilization in terms of sensory and nutritional quality compared to conventional retorting were demonstrated. Microwave heating models to predict the heating effects of food have for 15–20 years developed into increasingly more realistic models by numerical modelling (Wäppling Raaholt and Waldén, 2011; Wäppling Raaholt, 2000). Validation of such models is performed by experiments, where real product temperature distributions are measured or established. This is particularly important in pasteurization or sterilization applications.

Among industrial examples of microwave pasteurization of foods are pre-packed foods like pouch-packed meals or yoghurt (Rosenberg and Bögl, 1987b; Decareau, 1986). For several years, plants for commercial pasteurization of ready-made meals (Figure 18.3) are being offered by different companies and installations are increasingly spreading worldwide. Industrial microwave plants have been in commercial use for many years for pasteurization of packaged bread, cakes and confectionery, especially in countries where chemical mould inhibitors are not permitted or where their effect on the bread volume and aroma is not deemed acceptable.

Applications for microwave *sterilization* of packaged foods have been investigated for many years and were reported for packaged, pre-cooked foods by the US Army as early as 1971. At present, it has also been used commercially (e.g. Top's Foods in Europe, http://www.topsfoods.com). The requirements for temperature control are even more stringent for sterilization, which makes the need of process validation, assisted by advanced modelling tools to predict 'cold spots', particularly apparent.

Pasteurization and sterilization of fluid and semi-fluid foods The application of microwave pasteurization of fluid and semi-fluid foods is promising and gives several advantages (Wäppling Raaholt, Isaksson and Hamberg, 2013; Wäppling Raaholt, 2012a, 2011a; Wäppling Raaholt, Isaksson and Janestad, 2011; Isaksson, 2002; Ohlsson, 1993). The pumpable food is microwave heated across the tube cross-section during transportation through the tube. Uniformity of heating requires an appropriate design of the system, including

applicators, cavities and tube dimensions (Wäppling Raaholt, Isaksson and Janestad, 2011; Isaksson, 2002; Ohlsson, 1993). Systems for continuous fluid pasteurization or sterilization have been successfully developed (Wäppling Raaholt, Isaksson and Hamberg, 2013; Wäppling Raaholt, 2012a, 2011a; Isaksson, 2002; Ohlsson, 1993). While pasteurization can occur at atmospheric pressures, sterilization requires higher temperatures, and therefore increased pressure, in order to achieve satisfactory short sterilization times and to maintain high product quality. Microwave pasteurization and sterilization offer very rapid heat processing, which should lead to only small quality changes due to thermal treatment according to the HTST principle. However, the requirements on heating uniformity are very high in order to fulfil the quality advantages (Ohlsson, 1991). It is critical to know and control the lowest product temperatures, where the inactivation of microorganisms has the slowest rate.

There are also systems where the food is transported through the heating zone by a screw (Berteaud, 1995). Dehghan *et al.* (2012) concluded that microwave pasteurization is a good alternative to HTST pasteurization, based on a study on physicochemical characteristics in terms of protein, fat, acidity and solubility percentages due to heat treatment with either microwave pasteurization or HTST sterilization. Furthermore, the same study concluded that the contents of six amino acids and fatty acids did not differ between the two heating technologies. There are several studies reporting quality advantages of pumpable foods that were continuously microwave pasteurized (Wäppling Raaholt, Isaksson and Janestad, 2011; Lin and Ramaswamy, 2011) or sterilized (Wäppling Raaholt, Isaksson and Hamberg, 2013; Wäppling Raaholt, 2012a). The technique of continuous microwave sterilization of pumpable foods was demonstrated at SIK in Sweden for different kinds of foods, including particulate food systems (e.g. soups or sauces with pieces of vegetables and fish or meat). Figure 18.4 shows dissipated power in the load of a tubular microwave sterilization process. In the process, the pumpable food passes through one centre-heating (a) and two periphery-heating (b and c) cavities. Since the temperature of the food typically undergoes a dramatic change in a short distance in such a process, it is of interest to involve the temperature dependence of the dielectric properties, which is illustrated by the nonsymmetric power distribution in Figure 18.4. The direction of movement of the load is from left to right in each cavity/subplot.

The technology has also been studied for continuous pasteurization of fluids like milk (Rosenberg and Bögl, 1987b; Decareau, 1986), coagulation of liver paste (Isaksson, 2002) and low-acid multiphase products (Kumar *et al.*, 2007). However, a commercial application (not based on the same microwave mode of excitation of the cavities) is available in terms of the production of sweet potato puree (microwave sterilization followed by aseptic packaging) (Coronel *et al.*, 2005).

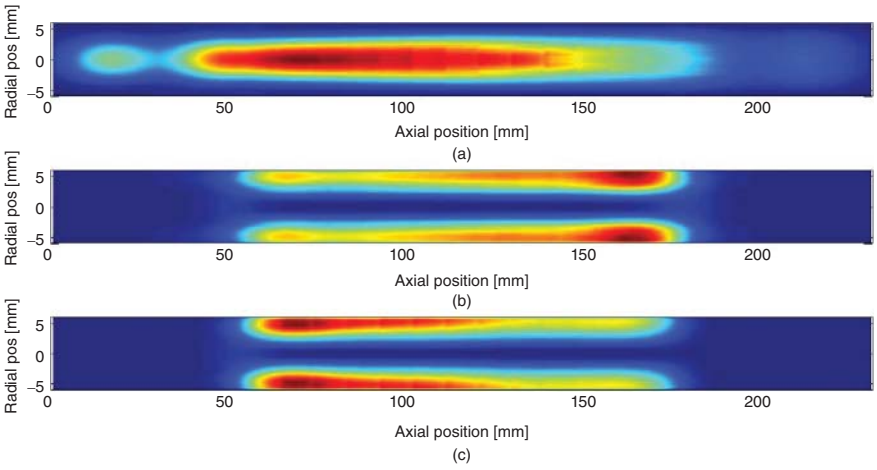


Figure 18.4 Dissipated power in the load of a tubular microwave sterilisation process (pos stands for position). (source: Isaksson 2013. Reproduced with permission of SIK, Sweden) *See plate section for colour version*

18.4.2 Tempering and defrosting

Microwave tempering of frozen food blocks is widely used in the food industry all over the world, mainly for tempering of meat, fish and butter (Ohlsson and Bengtsson, 2001; Bengtsson, 2001). A figure of 250 units globally is mentioned by Metaxas (1996).

Tempering means that frozen foods are thermally treated to increase the temperature from below −18 °C to temperatures just below the melting point of ice (approximately to −2 °C in foods of normal salt content). This increase of temperature results in mechanical product properties that are better suited for further machining operations such as cutting or milling. Conventional tempering requires large storage rooms and gives a significant drip loss and risk of microbial growth. By using microwaves, most often at the frequency of 915 MHz (896 MHz in the UK), the tempering time can be reduced to minutes or hours and the required space is reduced to one-sixth that of a corresponding conventional system (Metaxas, 1996). The possibility to use microwaves at low air temperatures gives another advantage in terms of reducing microbial growth. In order to avoid localized melting, which would correspond to a thermal run-away effect, it is important to control heating uniformity at the end temperature.

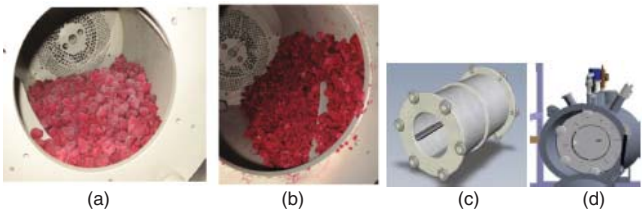
18.4.3 Drying and puffing

Drying is one of the most energy-consuming unit operations in the food industries. For this reason, microwave drying is a technology of interest, as the

efficient, in-depth heating leads to increased drying rates and shorter drying times, which result in lower energy consumption (Holtz *et al.*, 2009; Sharma and Prasad, 2002; Tulasidas, 1995; Tulasidas, Raghaven and Mujumdar, 1995). Other advantages that result from the shorter drying times are less material tied up in production, shorter drying tunnels, less space requirement in the processing plant, as well as the potential to improve quality, such as less shrinkage, better colour and better rehydration properties, as well as a better nutritional value of the dried products (Jaroenkit, Matan and Nisoa, 2013; Ji *et al.*, 2012; Arikan *et al.*, 2012; COMBIDRY, 2005).

The advantages of using microwave heating in drying are especially large when drying porous solid materials, as these materials are in general poor heat conductors and heat transfer using conventional methods is slow, especially in the partly dried materials. Microwaves are, therefore, often combined with other methods used for solid porous foods such as convective air drying, vacuum drying and freeze drying as they are all relatively time-consuming. In the area of food drying, microwave-convective drying, also called microwave-assisted drying, has been used commercially for drying sugar cubes, potato slices, pasta, onions and vegetable soup ingredients, and for puffing of snacks and baking or post-baking thin goods such as biscuits (Bengtsson, 2001; Decareau, 1985; Metaxas and Meredith, 1983). In microwave-vacuum drying, the reduced pressure limits the product temperatures as long as enough free water is present. The use of vacuum contributes to retaining product quality and more rapid drying. The high drying rate also allows for retention of water-insoluble aroma and leads to less shrinkage (Ehrle, 2005). Commercial applications of microwave-vacuum dehydration include fast drying of grains without germination (Decareau, 1985). By combining microwave-vacuum drying and air drying, puffed dried fruits and vegetables can be produced. Puffing of food products like fruits, vegetables and starchy products such as rice can be achieved by applying microwave heat to create a fast vaporization, which generates an open pore structure (Zheng *et al.*, 2013; Mohapatra and Das, 2011; Argyropoulos, Heindl and Müller, 2011). After puffing, the water content is further reduced to create a dried product. Often a combination of microwave and vacuum drying is used to create puffing. Some products are first stabilized through case hardening by pre-drying with conventional air drying. A pre-drying stage with air drying can also be used to produce dried fruits and vegetables, with improved rehydration properties (Räuber, 1998). Combinations of microwave and other drying techniques can also be used to create puffing. Microwave puffing is used industrially for the preparation of several types of puffed snack foods from cereals or grains.

Microwave-assisted freeze drying, as described several years ago by Sunderland (1980), has now moved to industrial implementation. Commercially available equipment is exemplified in Figure 18.5 (Püschner, 2013).



Brix	8–13%
Initial weight	5.kg (83% moisture content)
End weight	0.82 kg (7–10% moisture content)
Drying time	3.4 hours
Max temperature	35°C
Applied micro wave energy	4.64 kW

Figure 18.5 Microwave freeze dryer (a), (b), based on a closed rotating drum (c), (d), for preservation of raspberries (source: Püschner 2013. Reproduced with permission of Püschner.) *See plate section for colour version*

The dielectric properties are strongly influenced by temperature and moisture content, as well as other factors such as salt and sugar content, and will change substantially during drying of a material. The properties that govern the absorption of microwave energy have a large impact on the drying process, but are not alone sufficient to predict the temperature development and drying kinetics. There are also other material characteristics that change during drying, such as transport properties, heat capacity and, for hygroscopic materials, the energy required to evaporate the more tightly bound water in the material (Holtz *et al.*, 2010). The temperature profile induced by the microwave field will create a pressure profile, which in turn creates a moisture profile. The changes in moisture content will change the dielectric properties that alter the field pattern and the temperature profile. This complex process, coupled with the lack of material data for a wide range of temperature and moisture content, makes microwave-assisted drying difficult to predict. Turner and Jolly (1990) have indicated that materials may behave differently during drying depending on whether the dielectric properties are temperature and/or moisture dependent.

18.4.4 Pre-cooking, cooking and coagulation

Pre-cooking processes can also be accelerated by means of microwaves, as has been established for pre-cooking of poultry (Decareau, 1986), meat patty and bacon. Two main advantages are time reduction and improved yield. In bacon pre-cooking, microwave processing gives an important advantage in product appearance. It also diminishes the risk of nitrosamine formation and results in greater stability and higher quality of the rendered fat. In pre-cooking bacon, convective air flow removes the surface water while microwaves are used as the main energy source. In this way, coagulation of proteins and rendering of fat are performed by increasing the temperature. This process also gives a valuable by-product, rendered fat of high quality,

which could be used as a food flavourant (Ohlsson and Bengtsson, 2001; Schiffman, 1986). Microwave/steam cooking of chicken has received renewed attention with the advent of improved processing equipment and increasing market recognition of the gains possible in yield and sensory quality.

Baking Microwave technology is mainly used to accelerate baking, resulting in reduced processing times and an enhanced throughput. Reduced processing time could also correspond to space-saving equipment. Such accelerated baking could be achieved without loss of product quality and could be combined with convection or infrared heating to allow for colouring, crust formation and surface browning (Rosenberg and Bögl, 1987a). Microwave heating inactivates enzymes fast enough (due to a fast and uniform temperature increase throughout the product) to prevent starch from extensive breakdown, while developing sufficient carbon dioxide and steam to produce a uniform porous structure (Decareau, 1986). Recently, microwave baking has been successfully demonstrated in combination with colouring by means of an appropriate combination of infrared heating and convection, for both rolls and tin loaves (Wäppling Raaholt, 2012b), with advantages including reduced baking time (one-third of the time used for conventional baking), good product quality and less space requirements. Today, the main use of microwave baking is microwave finishing. Conventional baking is then used at the beginning with a high moisture-containing dough, while microwaves are used to improve the end of the baking process; the low heat conductivity would otherwise have resulted in long baking times. The application of microwave baking of crustless bread has also been industrially implemented with products sold both in Europe and globally. Microwaves can also be applied for microwave-assisted frying of doughnuts (Schiffman, 1986), leading to a shorter frying time and a lower fat uptake. Microwave proofing of dough is another potential application (Batey *et al.*, 1981) by combining partial proofing in a conventional proofer, followed by microwave proofing equipped with a warm, humidity-controlled air proofing system; the time could be reduced by 30–40%.

Blanching Since microwave heating of foods is performed by means of volumetric heating, microwave blanching does not require any water or steam. The rapid volumetric heating could also lead to reduced energy consumption as compared to conventional blanching.

The applications of microwave blanching of fruits and vegetables, either by means of microwaves alone or in combination with steam or water heating, as well as with different drying techniques, have been reported (Dorantes-Alvarez *et al.*, 2011; Ramesh *et al.*, 2002). Ramesh *et al.* (2002) evaluated the effect of microwave blanching on product properties (size, thickness and shape) and also compared pulsed microwave blanching with conventional water blanching at 95 ± 2 °C. The results showed that

microwave blanching resulted in more nutritious vegetables than those heated to the same temperature by water blanching. The results also indicated that microwave blanching was comparable to water blanching in terms of peroxidase inactivation. A study by Dorantes-Alvarez *et al.* (2011) concluded that microwave blanching enhanced antioxidant activity of peppers, *Capsicum annuum* Jalapeño, when microwave heat treatment was used to inactivate polyphenol oxidase. Industrial applications of this technology include microwave blanching of corn, potato and fruit (Ohlsson and Bengtsson, 2001; Decareau, 1986).

Re-heating of ready-to-heat meals Ready-to-heat meals for consumer microwave ovens can be developed into products with uniform microwave heating. By means of modelling-based design, where the microwave heating distribution in the food item is predicted by modelling of the electromagnetic fields, it is possible to select an appropriate combination of geometry, size and placement of the food components, as well as a suitable geometry of the food packaging. It is important that ready-to-heat meals, which are heated uniformly in the microwave oven, meet the expectations and needs of the customers. The interested reader can find more about the methodology in the references (Wäppling Raaholt, 2000; Wäppling Raaholt and Ohlsson, 2000).

18.4.5 Other microwave food processing applications

Edible fat can be obtained from animal by-products, for example from chicken skins or from abdominal fat. Microwave heating can result in high fat yields of good quality compared to other methods (Sheu and Chen, 2002). Similarly, microwave processing is also applied industrially for melting of chocolate, lipids, oil/fat and ice cream (Metaxas and Meredith, 1983). Microwave roasting is another example where the technology is applied industrially, for exampl for products like coffee beans, cocoa beans and nuts (Decareau, 1986), and also for roasting of beef (Metaxas and Meredith, 1983).

There are several applications where microwaves are combined with conventional technologies. Microwave-infrared baking of bread is one such example, which has shown successful baking of high-quality bread products with a much reduced baking time and energy consumption (Wäppling Raaholt and Isaksson, 2013; Wäppling Raaholt, 2012b, 2011b). Microwave-vacuum and microwave-freeze drying are other examples, where food quality is enhanced considerably, for example in terms of retention of temperature-sensitive substances like vitamins and colours. Furthermore, less shrinkage during microwave-vacuum drying as compared to conventional drying has been reported (Ehrle, 2005).

Thus, the main specifications needed for food industries regarding microwave equipment are complex and each specific application will have specific requirements depending on the product to be manufactured. In

addition, in many applications better results, in terms of energy consumption or/and product quality, are obtained by combining microwave with other heating methods, which further complicates equipment design.

18.5 Present status and future possibilities

18.5.1 Bottlenecks

In food industries, it is extremely important that production is not disturbed by production stops, food quality is uniform and safety levels are high. These issues are often pointed out as more important than short process time, low production costs or improved product quality. Cleanability of the equipment is an important issue, as well as good integration of the new equipment in the existing production flow.

For fast industrial implementation of new technologies, it is necessary to have 'success stories' to be demonstrated as examples. Concrete advantages and production sites are essential. This applies to any new technology.

Another hindrance has been the fact that food industry suppliers are often sector specific, and the food industry has specific long-term collaboration and agreements with equipment suppliers, making it difficult for new suppliers of equipment to penetrate into the market. Lack of knowledge of microwave suppliers about materials, cleaning/disinfection and understanding of the sector needs is an important obstacle.

In addition, the food industry is conservative and would not like to change a product recipe in order to implement a new process better. The food industry can also demand exclusive rights for a given application, which makes it difficult for equipment suppliers to further develop their business.

The typical company commercializing microwave equipment is small and may not have the economic capacity to build a large microwave equipment due to difficulties in getting capital and the high risk of bankruptcy in case component suppliers do not supply or equipment does not work as planned.

According to Bengtsson (2001), the main reasons why many microwave food applications have not been successful in the 1970s and 1980s are:

- Food products are complex and food production is regulated by authorities. Equipment suppliers have limited knowledge about the needs and requirements for food production and food processing, and the food industry has limited knowledge about microwave technology. Collaboration between both parts was difficult, resulting in bad performance of the equipment and increasing distrust against the technology among factory personnel.
- Product throughput is very high in the food industries; most processing systems operate at 500 to 5000 kg/h. Therefore, microwave equipment must also be able to operate at this capacity to enable integration in

existing production lines. There are systems that could meet the capacity requirements, but some systems exist that do not have the required capacity. An example is pasteurization of juices, which worked well in the lab, but scaling up to typical throughputs of thousands of litres per hour failed due to technical and economic reasons.

- Economic evaluation has not been well done. The intrinsic value of foods is low, while microwave processing costs may sometimes be relatively high – at least if the payback time is not taken into account. As a rule of thumb, the intrinsic value of food must be at least 2–4 dollars/kilogram to even consider its adoption. In the US, any addition of cost to the manufacture of a product is multiplied by 4 or 5 in its final retail price. In other words, if a microwave process adds $0.02 to the manufacture costs, the selling price will increase by $0.10. Unless the market is exclusively controlled by the producer, such a price increase may be difficult to implement if not significant improvements in product properties are achieved.
- Evaluation of microwave processing in comparison with traditional processing has not been well performed. Microwave processing may simplify raw material handling, for example microwave drying of chips instead of frying, as well as bringing other benefits that should be considered (microwave drying could have health benefits over frying due to reduced fat content of the final product, etc.).
- Food processors are rarely willing to change a product's formulation. Microwave processing may for some applications work better if small formulation changes are made.
- The definition of the processing goal for the microwave equipment was unclear and, for some equipment, there was a lack of process control.
- Microwave technology performed well but there were difficulties in integration of the process on their production line.
- Technology gave the expected results, but equipment breakdowns and other technical problems made the operators suspicious.

However, the number of successful installations has grown rapidly during the last two decades. One contributing factor, suggested by Ohlsson and Bengtsson (2001), is that the use of modelling tools to enable proper design of microwave equipment has made it possible to develop and construct optimal microwave ovens and equipment that ensures a uniform microwave heating distribution for several applications of interest.

When it comes to necessary factors to succeed in implementing microwave technology into cost-effective industrial applications, the following contributing factors are mentioned (Bengtsson, 2001):

- Applications need to enhance the unique advantages of microwave processing, such as penetration depth, accelerated heating rate and flexibility, which traditional technologies cannot offer.
- Reliability has to be high and process capacity adapted to the production capacity.
- Microwave technology needs to be energy effective. In some cases, microwave processing can be combined with traditional heating methods to achieve an energy effective and more uniform heating.
- A more effective collaboration between microwave specialists, equipment suppliers and food technologists is required.
- Microwave processing must simplify processing and reduce production costs and/or improve production capacity.
- High-quality production yield.
- Time for implementation of the technique in the industry is right both for management, technical personnel and operators.
- Improved functionality, safety and service.

18.5.2 Future trends

Industrial microwave processing has been considered very promising for decades, but practical application has, especially between 1960 and 1980, been relatively slow to develop. Several factors now point towards accelerated growth for several years to come:

- The reliability of microwave processing equipment is nowadays well comparable to that of alternative processing equipment, which makes the advantages of microwave heating more attractive. The operating life of industrial microwave equipment is today much longer, due to the development of more reliable magnetrons and the invention of ferrite circulators, which protect the generator tubes.
- The steadily reduced cost of microwave equipment in the last few decades is now comparable to that of conventional heating equipment.
- The future market for microwave processing equipment is promising for several reasons (e.g. shorter process time and much improved product quality, increased flexibility, reduced energy consumption in several application examples and less space requirements in production). Furthermore, it relies on the interdisciplinary understanding of the physical phenomena behind microwave heating, governed by Maxwell's equations, the interaction of the fields and the food to be heated, and also the dissipation of the microwave energy as heat, described by the electromagnetic equations as well as heat and mass transfer phenomena within the food during heating.

Prediction of the microwave heating distribution in terms of microwave modelling gives a powerful tool to optimize a microwave process system.

- The strong trend in today's food industry towards continuous processing lines and on-line process control indicates that there will be a growing need for microwave heating as a unit operation for very rapid and in-depth heating.

In the future, the dominating industrial applications will probably continue to be tempering, pasteurization, drying and production of snacks, but with a steadily growing number of installations for baking and sterilization. For industrial processing, it would be very important that the frequency of 915 MHz could be generally recognized as an ISM frequency, due to its advantages in larger penetration depth, larger generator power and efficiency.

When it comes to the market for domestic microwave ovens, it will presumably continue to grow worldwide, even if it has reached a near-saturation point in a few countries. The proportion of combination ovens, which combine microwaves with heat by convection or radiation, will continue to grow, at least in Europe.

Packaging development has previously focused on appropriate materials, shape, shielding, susceptors and other forms of active packaging. This development is expected to continue, at the same time as product development more and more takes into account the permittivity and thermal properties of foods and their interaction with the electromagnetic fields. Tailor-made aluminium foil susceptors and shielding were introduced in the market several years ago. They are expected to give a substantial increase in consumer satisfaction with the microwaveable food products. Further development may be required in the future to reduce the environmental impact of the use of resources for producing packaging material.

In research, further development of mathematical modelling and simulation tools will be invaluable in order to optimize process design of microwave plants and ovens, to evaluate performance and costs, and to reduce the need for empirical and experimental work. Modelling tools will become even more sophisticated and will continue to be important in studies of the influence of oven or applicator design on the field distribution and the resulting heating patterns in foods, and in evaluating the role of food shape and composition. It will also to a large extent facilitate studies on combinations of microwave heating with other forms of heating.

18.6 Conclusions

In the 1960s, food industries and equipment manufacturers predicted an exceptional future for industrial microwave processing, while very few expected that household microwave ovens would become popular. In early 2000, the situation was quite different. The household microwave oven

production rate was about 25 million units/year, with an estimated amount of 200 million units installed, while less than 500–1000 industrial microwave processing units were installed. How could the experts be so wrong? The number of successful industrial microwave installations has started to grow during the last decade, but why did it take so long to break the trend?

In food industrial operations continuous processes are usually needed, due to the desired high throughputs. Thus, microwave continuous operations need to be developed. The industrial microwave ovens may be categorized into high-power single-magnetron and low-power multimagnetron devices. Microwave applicators may have different configurations and are designed based on the properties, shape/size and volume of the material being heated. The design of applicators for food applications is complex and needs to be done with proper care, taking regard of the interaction between parameters important for heating uniformity, such as dimensions of cavity, applicators or waveguide system, food composition, size and geometry, packaging geometry and composition.

Movements of the product to be heated and/or wave stirrers are also used to improve heating uniformity, as well as control of the microwave power during processing. Conveyor belt ovens have been developed that are specifically for microwave treatment. The openings for the continuous ovens have to be properly designed in order to avoid leakage radiation, which is limited in the US and Europe by law to 5 mW/cm^2.

References

Argyropoulos, D., Heindl, A. and Müller, J. (2011) Assessment of convection, hot-air combined with microwave-vacuum and freeze-drying methods for mushrooms with regard to product quality. *International Journal of Food Science and Technology*, **46**(2), 333–342.

Arikan, M.F., Ayhan, Z., Soysal, Y. and Esturk, O. (2012) Drying characteristics and quality parameters of microwave-dried grated carrots. *Food and Bioprocessing Technology*, **5**(8), 3217–3229.

Balanis, C.A. (1989) *Advanced Engineering Electromagnetics*. John Wiley & Sons, Inc., New York, USA, 353 pp.

Batey, R.W., Grillo, R. J., Mirman, A.H., Schiffmann, R.F. (1981) Microwave baking with metal pans. US Patent US4388335 A.

Bengtsson, N. (1971) Dielektrisk värmning inom livsmedelstekniken – mätning av livsmedels dielektriska egenskaper samt försök med upptining och pastörisering. In: *Avhandling för Tekn*. Doktorsexamen, Institutionsavdelningen för Livsmedelskemi, CTH och Svenska Institutet för Konserveringsforskning (SIK), 112 sidor.

Bengtsson, N. (2001) Development of industrial microwave heating of foods in Europe over the past 30 years. *Journal of Microwave Power and Electromagnetic Energy*, **36**, 227–240.

Bengtsson, N. and Risman, P.O. (1971) Dielectric properties of foods at 3 GHz as determined by a cavity perturbation technique. *II. Measurements on food materials. Journal of Microwave Power*, **6**(2), 107–123.

Berteaud, A.J. (1995) Thermo-star. Bulletin from MES, 15 Rue des Solets, Rungis, France.

Buffler, C.R. (1993) Microwave Cooking and Processing. In: *Engineering Fundamentals for the Food Scientist*. AVI, van Nostrand Reinhold, New York, USA, 169 pp.

Cheng, D.K. (1989) *Field and Wave Electromagnetics*, 2nd edn:, Chapter 8: Plane electromagnetic waves; Chapter 10: Waveguides and cavity resonators. Addison-Wesley.

COMBIDRY (2005) New combined drying technologies for development of high quality shelf-stable fruit products. Final Report, ICA4-CY-2002-10034, International Scientific Cooperation Projects (1998–2002).

Coronel, P., Truong, V.-D., Simunovic, J. and Sandeep, K.P. (2005) Aseptic processing of sweet potato purees using a continuous flow microwave system. *Journal of Food Science*, **70**(9), E531–E536.

Decareau, R.V. (1985) *Microwaves in the Food Processing Industry*. Academic Press Inc., Orlando, Florida, USA.

Decareau, R.V. (1986) Microwave food processing equipment throughout the world. *Food Technology*, **40**, 99–105.

Dehghan, A., Jamalian, J., Farahnaky, A., Mesbahi, G. and Moosavi-Nasab, M. (2012) The effect of microwave pasteurisation on some physical and chemical characteristics of Milt. Int. *Journal of Food Engineering*, **8**, 1–12.

Dorantes-Alvarez, L., Jaramillo-Floresa, E., González, K., Martinez, R. and Parada, L. (2011) Blanching peppers using microwaves. *Procedia Food Science*, **1**, 178–183.

EC (2000) Food Project Results. AIR Agro-Industrial Research, EUR 19070, Contract AIR2-CT93-1054. In: A. Luchetti (ed.), Microwave sterilisation process validation nutritional wholesomeness, product quality and the consumer, pp. 40–43.

Ehrle, U. (2005) Drying using microwave processing. In: *Microwave Processing of Foods*, Schubert, H. and Regier, M. (eds). CRC Press, Boca Raton, Florida, USA, and Woodhead Publishing, pp.142–152.

Gabriel C., Chan, T.Y.A. and Grant, E.H. (1994) Admittance models for open ended coaxial probes and their place in dielectric spectroscopy. *Physical and Medical Biology*, **39**, 2183–2200.

Gabriel, C., Grant, E.H. and Young, I.R. (1986) Use of time domain spectroscopy for measuring dielectric properties with a coaxial probe. *Journal of Physics E: Science Instruments*, **19**, 843.

Goedeken, D.L., Tong, C.H. and Virtanen, A.J. (1997) Dielectric properties of a pre-gelatinized bread system at 2450 MHz, as a function of temperature, moisture, salt and specific volume. *Journal of Food Science*, **62**(1), 145–149.

Harrington, R.F. (1961) *Time-Harmonic Electromagnetic Fields*. McGraw-Hill Book Co., New York, USA, pp. 158–163.

Hasted, J.B. (1973) *Dielectrics and Waves*. John Wiley & Sons.

Hecht, E. (1989) *Optics*, 2nd edn, Chapter 7. Addison-Wesley.

Holtz, E. and Raaholt, B. (2013) Image remodelled from Innovation Guide, ITP portal, HighTech Europe, www.hightecheurope-portal.eu.

Holtz, E., Ahrné, L., Karlsson, T.H., Rittenauer, M. and Rasmuson, A. (2009) The role of processing parameters on energy efficiency during microwave convective drying of porous materials. *Drying Technology*, **27**, 173–185.

Holtz, E., Ahrné, L., Rittenauer, M. and Rasmuson, A. (2010) Influence of dielectric and sorption properties on drying behaviour and energy efficiency during microwave

convective drying of selected food and non-food inorganic materials. *Journal of Food Engineering*, **97**, 144–153.

Incropera, F.P. and Dewitt, D.P. (2002) *Fundamentals of Heat and Mass Transfer*, 5th edn. John Wiley & Sons, Inc., New York, USA, p. 981.

Isaksson, S. (2002) Tubular microwave heating. Thesis for the Degree of Licentiate of Engineering, Chalmers University of Technology, Sweden.

Jaroenkit, P., Matan, N. and Nisoa, M. (2013) Microwave drying of cooked brown rice and the effect on the nutrient composition and trace elements. *International Food Research Journal*, **20**, 351–355.

Ji, H., Du, A., Zhang, L., Li, S., Yang, M. and Li, B. (2012) Effects of drying methods on antioxidant properties and phenolic content in white button mushroom. *International Journal of Food Engineering*, **8**(3), DOI: 10.1515/1556-3758.2491.

Katt, J.L. (1991) The effects of starches and sugars on microwave cooking. *Microwave World*, **12**(2), 19–23.

Kent, M. (1987) Electrical and dielectric properties of food materials. In: *Bibliography and Tabulated Data*, Compiled by M. Kent, COST90bis, Science and Technology Publishers Ltd., Essex, England.

Kumar, P., Coronel, P., Simunovic, J. and Sandeep, K.P. (2007) Feasibility of aseptic processing of a low-acid multiphase food product (salsa con queso) using a continuous flow microwave system. *Journal of Food Science*, **72**(3), E121–E124.

Lin, M. and Ramaswamy, H. (2011) Evaluation of phosphate inactivation kinetics in milk under continuous flow microwave and conventional heating conditions. *International Journal of Food Properties*, **14**(1), 110–123.

Maxwell, J.C. (1954) *A Treatise on Electricity and Magnetism*, vol. 1-2, Unabridged version of the last third revised edition 1892. Dover Publications, New York, USA.

Metaxas, A.C. (1996) *Foundations of Electroheat – A Unified Approach*. John Wiley & Sons, Ltd, Chichester, England, 500 pp.

Metaxas, A.C. and Meredith, R.J. (1983) *Industrial Microwave Heating*. IEE Power Engineering Series 4, Peter Peregrinus Ltd, Piscataway, New Jersey, USA, 357 pp.

Mohapatra, M. and Das, S.K. (2011) Effect of process parameters and optimization on microwave puffing performance of rice. *Research Journal of Chemistry and Environment*, **15**(2), 454–461.

Mudgett, R.E. (1986) Electrical properties of foods. Ch 7. In: *Engineering Properties of Foods*, Rao, M.A. and Rizvi, S.S.V. (eds). Marcel Dekker Inc., New York, USA, pp. 32–90.

Mudgett, R.E. (1995) Electrical properties of foods. In *Engineering Properties of Foods*, 2nd edn, Rao, M.A. and Rizvi, S.S.H.(eds). Marcel Dekker Inc., New York, USA, pp. 389–455.

Mudgett, R.E., Goldblith, S.A., Wang, D.I.C. and Westphal, W.B. (1980) Dielectric behavior of semi-solid food at low, intermediate and high moisture contents. *Journal of Microwave Power*, **15**(1), 27–36.

Nelson, S.O. (1973) Electrical properties of agricultural products – a critical review. *Transactions of the ASAE*, **16**(2), 384–400.

Nott, K.P., Hall, L.D., Bows, J.R., Hale, M. and Patrick, M.L. (1999) Three-dimensional MRI mapping of microwave induced heating patterns. *International Journal of Food Science and Technology*, **34**, 305–315.

Ohlsson, T. (1981) Methods for measuring temperature distribution in microwave ovens. *Microwave World*, **2**(2), 14–17.

Ohlsson, T. (1983) Fundamentals of microwave cooking. *Microwave World*, **4**(2), 4–9.

Ohlsson, T. (1993) In-flow microwave heating of pumpable foods. Paper presented at the International Congress on *Food and Engineering*, Chiba, Japan, May 23–27, 7 pp.

Ohlsson, T. and Bengtsson, N.E. (1975) Dielectric food data for MW sterilisation processing. *Journal of Microwave Power*, **10**(1), 93–108.

Ohlsson, T. and Bengtsson, N.E. (2001) Microwave technology and foods. *Advances in Food and Nutrition Research*, **43**, 65–140.

Ohlsson, T. and Risman, P.O. (1978) Temperature distribution of microwave heating – spheres and cylinders. *Journal of Microwave Power*, **13**(4), 303–310.

Ohlsson, T. and Thorsell, U. (1985) Wiedererwärmen gekühlter Lebensmittel and Speisen mittels Mikrowellen *Ernährungs-Umschau*, **32**(4), 104–108.

Ohlsson, T., Bengtsson, N.E. and Risman, P.O. (1974) The frequency and temperature dependence of dielectric food data as determined by a cavity perturbation technique. *Journal of Microwave Power*, **9**(2), 129–145.

Püschner, P,-A. (2013) Presentation at the Seminar on *Microwave Applications*, KIN, Neumünster, 26 February 2013.

Ramesh, M.N., Wolf, W., Tevini, D. and Bognár, A. (2002) Microwave blanching of vegetables. *Journal of Food Science*, **67**(1), 390–398.

Räuber, H. (1998) Instant-Gemüse aus dem östlichen Dreiländereck. *Gemüse*, **10**.

Risman, P.O. (1992) Metal in the microwave oven. *Microwave World*, **13**(1), 28–33.

Risman, P.O. (1999) *Public Seminar at QuickWave Modelling Course*, Göteborg, Sweden, November 1999.

Risman, P.O. and Bengtsson, N. (1971) Dielectric properties of foods at 3 GHz as determined by a cavity perturbation technique. I. Measuring technique. *Journal of Microwave Power*, **6** (2), 101–106.

Risman, P.O., Ohlsson T. and Lingnert, H. (1993) *Model Substances and Their Use in Microwave Heating Studies*. SIK publication, SIK-rapport 588, 10 pp.

Rosenberg, U. and Bögl, W. (1987a) Microwave thawing, drying, and baking in the food industry. *Food Technology*, **41**, 85–91.

Rosenberg, U. and Bögl, W. (1987b) Microwave pasteurisation, sterilisation, blanching and pest control in the food industry. *Food Technology*, **41**, 92–121.

Ryynänen, S. and Ohlsson, T. (1996) Microwave heating uniformity of ready meals as affected by placement, composition and geometry. *Journal of Food Engineering*, **29**, 13–21.

Ryynänen, S., Risman, P.O. and Ohlsson, T. (2004) Hamburger composition and microwave heating uniformity. *Journal of Food Science*, **69** (7), M187–M196.

Schiffman, R.F. (1986) Food product development for microwave processing. *Food Technology*, **40**(6), 94–98.

Schiffman, R.F. (2001) Microwave processes for the food industry. In: *Handbook of Microwave Technology for Food Applications*. Marcel Dekker, Inc.

Schubert, H. and Regier, M. (eds) (2005) *The Microwave Processing of Foods*, CRC Press, Boca Raton, Florida, USA, and Woodhead Publishing, p. 17.

Sharma, G.P. and Prasad, S. (2002) Comparison of convective and microwave–convective drying of garlic: kinetics and energy consumption. *Journal of Food Science and Technology*, **29**(6), 603–608.

Sheu, K.S. and Chen, T.C. (2002) Yield and quality characteristics of edible broiler skin fat as obtained from five rendering methods. *Journal of Food Engineering*, **55**(3), 263–269.

Sundberg, M. (1998) An investigation of the edge overheating effect for high-permittivity dielectrics, Paper F. In: Analysis of industrial microwave ovens. PhD Thesis, Department of Microwave Technology, Chalmers University of Technology, Sweden.

Sunderland, J.E. (1980) Microwave freeze drying. *Journal of Food Processing and Preservation*, **4**(4), 195–212.

Tulasidas, T.N. (1995) Combined convective and microwave drying of grapes. Dissertation Thesis, UMI Dissertation Services, Michigan, USA.

Tulasidas, T.N., Raghavan, G.S.V. and Mujumdar, A.S. (1995) Microwave drying of grapes in a single mode cavity at 2450 MHz – II: Quality and energy aspects. *Drying Technology*, **13**(8 and 9), 1973–1992.

Turner, I. and Jolly, P. (1990) The effect of dielectric properties on microwave drying kinetics. *Journal of Microwave Power and Electromagnetic Energy*, **25**(4), 211–223.

von Hipple, A.R. (1954) *Dielectric Materials and Applications*. The Technology Press of MIT and John Wiley & Sons, Inc., New York, USA.

Walker, J. (1987) The secret of a microwave oven's rapid cooking is disclosed. *Scientific American*, (**2**), 98–102.

Wäppling Raaholt, B. (2000) Finite-Difference Time-Domain Analysis of Heating of Foods in Household Microwave Ovens. Thesis for the Degree of Licentiate of Engineering, Chalmers University of Technology, Department for Electromagnetics, Nov. 2000.

Wäppling Raaholt, B. (2009) Methodology for controlling the colour-shift temperature of a model food intended for microwave heating purposes. In: Proceedings of the 12th International AMPERE Conference on *Microwave and HF Heating*, Karlsruhe, Germany, September 2009, 4 pp.

Wäppling Raaholt, B. (2011a) Energieffektiv och innovativ tillverkning av livsmedel – med mikrovågsvärmning av pumpbara råvaror'', projekt delfinansierat av Jordbruksverket, slutrapport Dnr 19/8003-08 /P80633.

Wäppling Raaholt, B. (2011b) Energieffektiv bakning i liten skala, projekt delfinansierat av SJV, slutrapport Dnr 19/8003-08.

Wäppling Raaholt, B. (2012a) Snabb, skonsam och effektiv värmning med pumpbara råvaror'', projekt delfinansierat av SJV, delrapport 2012, Dnr 19/1325-11.

Wäppling Raaholt, B. (2012b) Energieffektiv bakning i pilotskala, projekt delfinansierat av SJV, delrapport 2012 Dnr 19/1324-11.

Wäppling Raaholt, B. and Isaksson, S. (2013) Combined microwave–infrared baking of bread. In: Conference Proceedings of the AMPERE Conference on *Microwave and High*-Frequency *Heating*, Nottingham, England, September 2013, pp. 35–39.

Wäppling Raaholt, B., Isaksson, S., Hamberg, L. (2013) A tubular microwave sterilisation or pasteurisation process for pumpable foods. In: Conference Proceedings of the AMPERE Conference on Microwave and High-Frequency Heating, Nottingham, England, September **2013**, pp. 80–83.

Wäppling Raaholt, B., Isaksson, S. and Janestad, H. (2011) Continuous microwave heating of pumpable foods – examples of two potential applications. In: The 13th International

AMPERE Conference on Microwave and HF Heating, Toulouse, France, September 2011, pp. 298–301.

Wäppling Raaholt, B. and Ohlsson, T. (2000) Tools for improving the heating uniformity of foods heated in a microwave oven. *Microwave World*, **21**(1), 24–28.

Wäppling Raaholt, B. and Ohlsson, T. (2005) Improving the heating uniformity in microwave processing. In: *Microwave Processing of Foods*, Schubert, H. and Regier, M. (eds). CRC Press, Boca Raton, Florida, USA, and Woodhead Publishing Ltd, pp. 292–316.

Wäppling Raaholt, B. and Ohlsson, T. (2009) *Influence of Food Geometry and Dielectric Properties on Heating Performance in Development of Packaging and Products for Use in Microwave Ovens*, Pesheck, P.S. and, Lorence, M.W. (eds). CRC Press, Boca Raton, Florida, USA, and Woodhead Publishing Ltd, pp. 38–65.

Wäppling Raaholt, B. and Risman, P.O. (2003) Permittivity of inhomogeneous food items by retro-modelling with a degenerate mode cavity. In: The 9th International AMPERE Conference, Loughborough, England, September 2003, 4 pp.

Wäppling Raaholt, B., Risman, P.O. and Ohlsson, T. (2001) Microwave heating of ready meals – FDTD simulation tools for improving heating uniformity. Presented at the 8th AMPERE Conference, Bayreuth, Germany, September 2001.

Wäppling Raaholt, B., Risman, P.O. and Ohlsson, T. (2006) Microwave heating of ready meals – FDTD simulation tools for improving heating uniformity. Presented at the 8th AMPERE Conference, Bayreuth, Germany, September 2001; also available as an extended paper (2006) in: *Advances in Microwave and Radio Frequency Processing*, Willert Porada, M. (ed.), Springer Verlag, pp. 241–255.

Wäppling Raaholt, B. and Waldén, B. (2011) Modelling of a combined microwave-infrared baking process of bread. In Conference Proceedings of the AMPERE Conference on Microwave and High-Frequency Heating, Toulouse, France, 2011, pp. 340–343.

Zheng, X., Liu, C., Shi, J., Xue, S., Mu, Y., Lin, Z., Liu, H. (2013) Analysis of volume expansion and dehydration rate of berry slab under microwave-vacuum puffing conditions. *LWT - Food Science and Technology*, **52**(1), 39–48.

19

Infrared in Food Preservation and Processing

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19.1 Introduction

Infrared (or radiant heat) is one of the most widely used heating techniques in various industries. Infrared (IR) radiation is the part of the electromagnetic spectrum that is predominantly responsible for the heating effect of sunlight (Ranjan, Irudayaraj and Jun, 2002). The infrared spectrum of radiation can be divided into three different categories (Figure 19.1), viz. near-infrared (NIR) radiation (wavelength: 0.75–3 μm), mid-infrared (MIR) radiation (wavelength: 3–25 μm) and far-infrared (FIR) radiation (wavelength: 25 – 1000 μm) (Sakai and Hanzawa, 1994).

Application of IR heat is one of the oldest ways to dry food products. Most applications of IR in food processing have been reported between 1950 and 1970. Later in 1970–1980, a considerable quantity of basic works on IR application in foods was conducted (Henry and Chapman, 2000). In food processing, the IR radiation band of 2.5 to 200 μm is mostly used (Nindo and Mwithiga, 2010). This chapter describes the theory and mechanism of IR heating and its application to different foods and processing industries.

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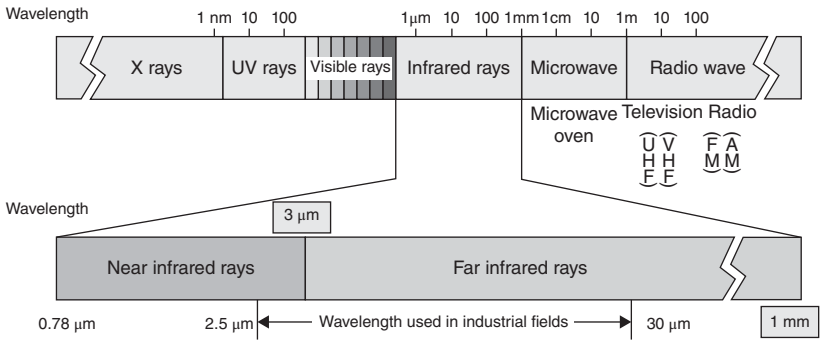

Figure 19.1 Electromagnetic spectrum showing the IR radiation band (source: Japan Far Infrared Association (JIRA): http://www.enseki.or.jp/e_ippo.html. Reproduced with permission of JIRA)

19.2 Theory of infrared drying

19.2.1 Mechanism of IR absorption by food

Radiant energy that falls on a food surface induces various changes in its components at molecular and atomic levels. The absorption mechanism is fundamentally governed by the spectrum of wavelength of the incident radiation. The main components of foodstuffs that absorb IR energy at wavelengths between 2.5 and 3.3 μm (medium- to long-wave IR) are water and various organic molecules, including protein and starch (Il'yasov and Krasnikov, 1991). The spectral bands and changes in state of atoms or molecules due to absorption of these radiation bands are given in Table 19.1. A major mechanism of absorption of IR by food occurs in the medium of the far-IR through stretching vibration of molecules that ultimately manifests as generation of heat in the product (Datta and Almeida, 2005). The approximate range related to strong absorption bands for major food constituents are 3–4 and 6–9 μm for proteins, 3–4, 6 and 9–10 μm for lipids and 3 and 7–10 μm for sugars (Figure 19.2). The four major absorption peaks of liquid water are 3, 4.7, 6 and 15.3 μm (Sandu, 1986). Radiation received by the food can be resolved into three parts, viz. absorbed radiation, transmitted radiation and reflected radiation (Figure 19.3). All these fractions depend upon the radiation properties of the food, which are defined as absorptivity (α), reflectivity (ρ) and transmissivity (τ). Thus, when IR radiation strikes the food surface, the thermal balance equation is given by

$$\alpha + \tau + \rho = 1 \tag{19.1}$$

Specifically, these properties are a function of the wavelength of the radiation. For monochromatic radiation, these are pre-fixed with a word 'spectral' and symbolized as α_λ, ρ_λ and τ_λ. In the case of polytropic radiation, they are defined as 'total' (Sandu, 1986).

Table 19.1 Changes in states of atoms and molecules in foods with absorption of different bands of radiation energy

Radiation	Wavelength range	Changes occur	Components involved	Reference
UV and visible	0.2–0.7μm	Electronic state	Atoms	Decareau, 1985
MIR and FIR	2.5 – 100 μm	Vibrational state	Molecules	
FIR	Beyond 100 μm	Rotational state	Molecules	

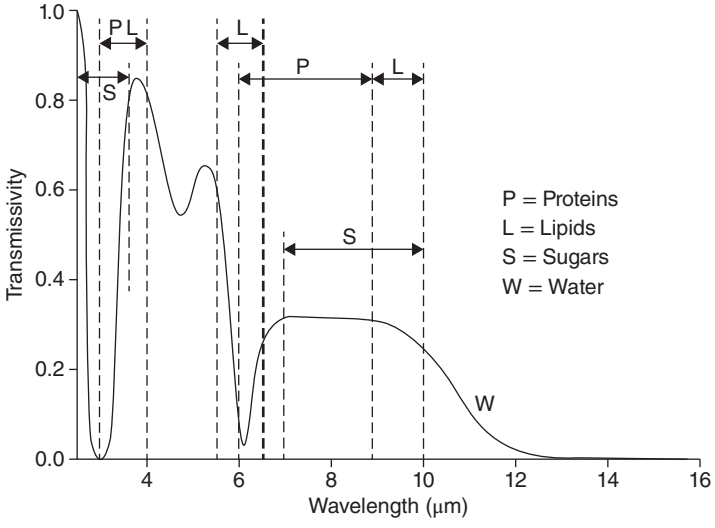

Figure 19.2 IR absorption bands of different components of food (source: Pan and Atungulu 2010. Reproduced with permission of Taylor and Francis Ltd)

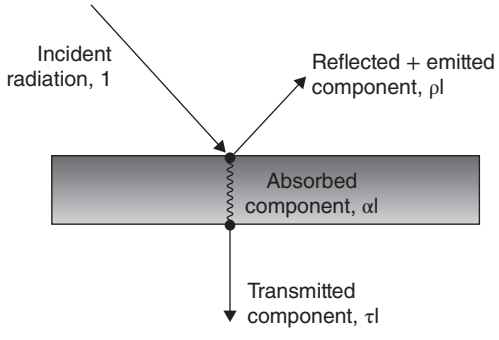

Figure 19.3 Propagation of radiation through the medium

Table 19.2 Depth of penetration of IR energy into different food products (source: Adapted from Pan and Atungulu. Reproduced with permission of Taylor and Francis)

Product	Spectral peak (μm)	Depth of penetration (mm)
Dough, wheat	1.0	4–6
Bread, wheat	1.0	11–12
Bread, biscuit	1.0	4
Dried bread	0.8	12
Grain, wheat	1.0	2
Carrots	1.0	1.5
Tomato paste	1.0	1
Raw potatoes	1.0	6
Dry potatoes	0.88	15–18
Raw apples	1.16	4.1
	1.65	5.9
	2.36	7.4

The depth of penetration of infrared radiation for different food products (Datta and Almeida, 2005) is shown in Table 19.2. This depth of penetration significantly influences both temperature and moisture at the surface of the products (Almedia, Torrance and Datta, 2006). IR radiation with less penetration power increases the surface temperature. In some food materials, short-wave IR has penetration depths of up to about 5 mm; this is almost ten times higher than that of long-wave IR. The latter has more effect on the surface.

19.2.2 Basic laws of radiation

Both spectral and directional features of IR radiation, like all other electromagnetic radiations, depend on temperature of the emitter and emissivity of the lamp. Three basic laws of black body radiation govern the relationship between the emissive power of the emitter, dominant wavelength of the emitted radiation and temperature of the emitter. These are Planck's law, Stefan–Boltzmann's law and Wien's displacement law (Sakai and Hanzawa, 1994; Dagerskog and Osterstrom, 1979).

Planck's law Spectral distribution of radiation energy from a black body at a particular temperature is governed by (Modest, 1993)

$$E_{b\lambda}(T, \lambda) = \frac{2\pi h C_o^2}{n^2 \lambda^5 [e^{hC_o/nk\lambda T} - 1]} \tag{19.2}$$

where k is Boltzmann's constant (1.381×10^{-23} J/K), n is the refractive index of the medium, λ is wavelength (m), T is the absolute temperature of the black body (K), C_o is the speed of light (2.998×10^8 m/s), h is Planck's constant (6.626×10^{-34} J s) and $E_{b\lambda}$ (T,λ) is the spectral distribution of radiation energy.

Stefan–Boltzmann's law This law states that the total energy emitted by a radiator depends upon the fourth power of its absolute temperature. Thus, the amount of infrared radiant energy from an emitter can be estimated from

$$E_b = \int_0^\infty E_{b\lambda}(T, \lambda)\, d\lambda = n^2 T^4 \int_0^\infty \frac{C_1 d(n\lambda T)}{(n\lambda T)^5 (e^{C_2/n\lambda T} - 1)} = n^2 \sigma T^4 \qquad (19.3)$$

where C_1 is $2\pi h {C_o}^2 = 3.75 \times 10^{-16}$ W/m^2, C_2 is hC_o/T μm K, σ is the Stefan–Boltzmann constant (5.670×10^{-8} W/m^{2}K^4) and E_b is the heat flux. The heat flux, estimated using this law, should be consistent with the amount of estimated heat flux for the entire spectral range using Planck's law (Equation (19.2)).

Wien's displacement law Wein's displacement law states that, corresponding to any absolute temperature (T) of the black body, maximum emissive power occurs at a particular peak wavelength (λ_{max}) and the product of λ_{max} and T bears a constant value (2898 μm K). Thus, the peak wavelength of radiation shifts towards a higher value (long-wave radiation) as the temperature of the body decreases (Figure 19.4) and vice versa.

19.2.3 IR emitters and spectral bands

In general, two types of infrared (IR) emitters are used in the industry. These are electric IR emitters and gas-fired IR emitters. Electric infrared emitters emit radiations with its wavelength varying between short and long waves, depending on the magnitude of the supply voltage. The gas-fired IR emitters, on the other hand, emit medium or long wavelength IR radiation. Short wavelength radiation (0.7 to 1.4 μm) corresponds to a source temperature around 1300–2600 K. Emitters in this group can deliver very high power density equal to or more than 300 kW/m^2, and attain a maximum temperature within a few seconds. Medium-wave infrared emitters take about a minute to emit radiation, with wavelengths ranging between 1.4 and 3.0 μm. This corresponds to a temperature range of 850–1200 K and power density of 90 kW/m^2. Medium wave IR emitters are extensively used for drying and curing of food products. Long-wave infrared emitters have a radiation temperature of 500–800 K, which corresponds to a spectrum of wavelength greater than 3.0 μm. It attains a power density up to 40 kW/m^2 (Table 19.3). Short-wave IR radiation can penetrate deep into the solid materials and

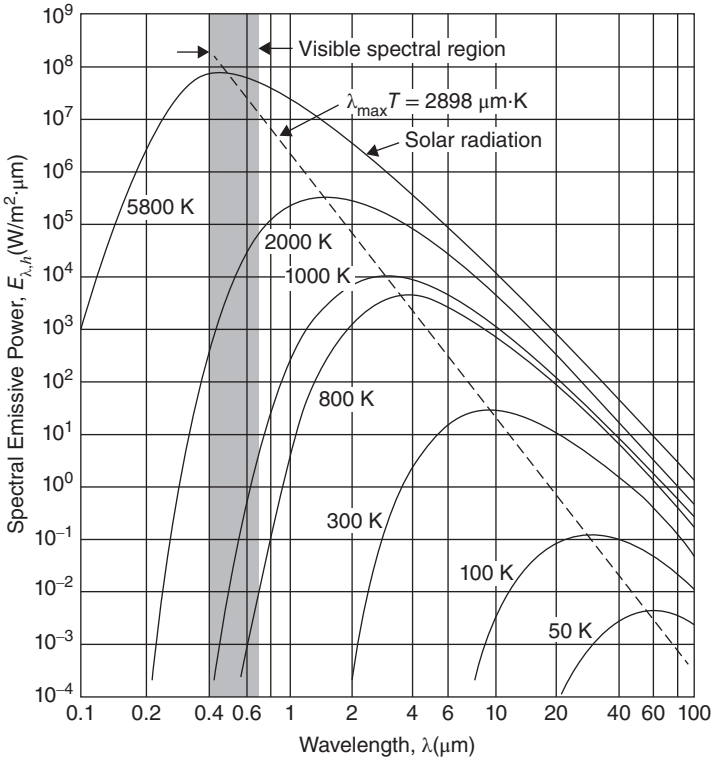

Figure 19.4 Spectral distribution of black body radiation at different temperatures (source: Incropera and Dewitt 1996. Reproduced with permission of John Wiley and Sons, Inc)

ensures uniform heating. Medium-wave radiation is absorbed generally at the surface and consequently its temperature rises with heating. Operating efficiency of electric IR heaters and gas-fired IR heaters varies between 40 and 70%, and 30 and 50%, respectively (Ramaswamy and Marcotte, 2005). Since the wavelength of the emitted radiation significantly influences the heating process, selection of the correct emitter becomes a very important factor. High efficiency in IR drying of foods could be achieved with specific IR generators. The peak wavelength of radiation with these emitters matches with the absorption bands of water in the foods. The uniformity and extent of absorption of IR energy also depends on size, shape and composition of the food materials; thus it becomes location specific in a composite food. Non-uniformity in radiation flux density on the surface of the irradiated foodstuffs is attributed to improper spacing between the consecutive emitters in the array (in a system of multiple emitters) and the gap between the emitter and the receiver (Il'yasov and Krasnikov, 1991).

Table 19.3 Comparison of different forms of infrared wave emitter (source: Adapted from Pan and Atungulu. Reproduced with permission of Taylor and Francis)

Parameters	Short-wave	Medium-wave	Long-wave
Infrared parameters	Halogen lamp	Quartz emitter	Resistance material
Material	Quartz sealed tube with tungsten coil inside	Quartz tube with Fe–Cr–Al alloy as heating element	Steel tube with Fe–Cr–Al alloy as heating element enclosed
Radiation efficiency	High: ~92%	Medium: ~60%	Low: ~40%
Peak wavelength	1.2 μm	2.2 μm	4.0 μm
Radiator temperature	2500 K	1300 K	800 K
Major heating mode	Radiation	Combined radiation and convection	Convection
Response time to 90% output	1 s	30 s	300 s
Convergence of radiation	Possible with good focusing	Fairly possible	Fairly possible

19.3 Application of infrared energy in food industry

Radiant heat transfer is often said to be more efficient and cost effective than convective heat transfer. It is largely because radiant heat transfer reduces cycle times, focuses energy on the target and produces virtually no volatile organic compounds, carbon monoxide (CO) and nitrogen oxides (NO_x) (Das and Das, 2010). Some other advantages of IR radiation are high heat transfer coefficients, a short process time, compact equipment and precise control over process parameters. Since air is transparent to IR radiation, the heating process can be done even at ambient air temperature, targeting directly the material without heating the intervening medium (Nowak and Lewicki, 2004).

Typically, in some cases, infrared (IR) radiation heating results in energy reduction as high as 245% compared to hot-air drying at 70 °C (Afzal, Abe and Hikida, 1999). Thus, the commercial importance of IR energy has been well understood by the food processing industry for its use in diverse sectors. Various thermal operations, viz. dehydration, blanching, baking, roasting, disinfestation, thawing and pasteurization, could use infrared radiation with success, replacing or in combination with conventional heating techniques. In some specific cases, IR radiation heating could also be applied as the finishing step for quality improvement of the product. Applications of IR radiation in specific food processing operations are presented in the subsequent sections.

19.3.1 Drying

Advantages in IR drying In the 1960s, applications of IR radiation in drying of agricultural and food products were envisaged along with establishing the related theory. In later years its applications in drying of grains, vegetables, fruits and other semi-processed food products were reported (Bal *et al.*, 1970; Hall, 1962; Schroeder and Rosberg, 1961; Yagi and Kunii, 1951). Versatile and simple constructional features of the drying system, convenience in heating using combinations of IR with conductive, convective and microwave heating, a fast heating rate and the preservation of flavour and other nutrients to a large extent are a few of the advantages in IR drying (Ranjan, Irudayaraj and Jun, 2002; Sandu, 1986).

Effect of depth and vibration Absorption of IR radiation by the product undergoing drying depends upon wavelength and intensity of radiation, moisture content, temperature and surface characteristics of the product. Typically, IR heating of wheat flour revealed that 99% of the absorbed energy remained concentrated in the surface layer up to a depth of 1/32 inch or less. This implies that IR drying can be suitably applied to thin-layer drying of agricultural products (Headly and Hall, 1963). In IR drying, moisture distribution varies widely along the depth of the grain bed (Person and Sorenson, 1962). The layer close to the emitter loses moisture more rapidly compared to that deep inside the bed. Adopting any mechanism for thorough mixing the grains in the bed (e.g. tumbling, turning or vibration) improves efficiency of drying with uniform absorption of IR energy and moisture distribution in the whole mass (Nindo, Kudo and Bekki, 1995). Vibration aided IR drying of a 12–16 mm paddy bed, receiving frequency of 20–22 Hz and with vibration amplitude of 8–9 mm (Figure 19.5) reported an insignificant effect of grain bed height on the drying rate (Das *et al.*, 2004, 2003). Table 19.4 gives concise information on IR drying of fruits, vegetables and nuts.

Direct and combined mode of heating In IR drying, product temperature and loss of some quality parameters, especially for fruits and vegetables, are directly proportional to the intensity of IR radiation. Many damages related to physical, mechanical and chemical properties (increased volume, lower rupture point and toughness, higher water uptake, higher leaching losses and reduction in trypsin inhibitor activity) are attributed to development of surface cracks in legume seeds (Fasina *et al.*, 2001) when the seeds are heated by IR to a surface temperature of 140 °C. The functional characteristics of flour from the infrared-heated seeds are superior to those of flour from untreated seeds. However, intermittent infrared heating and continuous convection drying of thick porous material resulted in good surface quality and energy efficiency (Dostie *et al.*, 1989). The combined system gives a

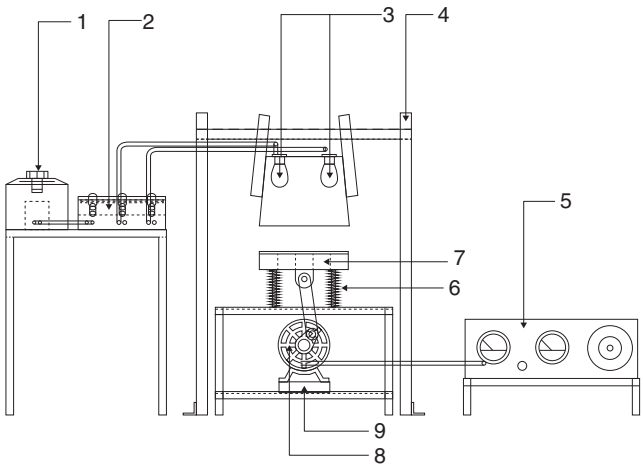

Figure 19.5 Schematic of vibratory batch type infrared dryer (1, variac; 2, wattmeter; 3, IR lamps; 4, main frame; 5, motor speed regulating unit; 6, helical springs; 7, drying tray; 8, motor; 9, base plate) (source: Das et al. 2004. Reproduced with permission of Elsevier)

synergic effect, as has been observed in barley drying in a thin layer under combined FIR–convection and convection alone (Afzal, Abe and Hikida, 1999). Far-infrared radiation enhances the drying rate with considerable reduction in energy consumption than at comparable convective temperatures. An increase in air velocity increased the specific energy consumption. A higher drying rate in thin-layer drying of onion slices, and consequently a reduced drying time, have been observed with an increase in infrared power at a given air temperature and velocity (Sharma, Verma and Pathare, 2005a, 2005b).

Better colour quality in drying of osmotically pre-treated vegetables and fruits (potato, carrot and banana) has been obtained under intermittent IR heating with a long tempering period and short exposure than samples dried by a pure convective condition (Chua *et al.*, 2004). Further, in comparison to constant IR intensity drying, step-down IR intensity (SDIRI) drying shortens drying time for these agro-samples compared to a longer drying time to step-up IR intensity (SUIRI) drying process; the colour change in dried samples is less with the latter technique than the former.

Combinations of IR radiation with other drying modes have also interesting results. Infrared (IR) radiation and freeze drying, either simultaneously or sequentially, are recent methods with considerable prospect for industrial applications. Freeze drying minimizes the negative impacts on final product quality attributes. A combination of IR and freeze drying systems have been evaluated with sweet potato (Lin, Tsen and King, 2005), strawberry (Shi *et al.*, 2008) and banana (Pan *et al.*, 2008).

Table 19.4 IR drying of fruits, vegetables and nuts

Product	Energy attributes	Drying attributes	Quality attributes	References
Onion slices	IR power – 300, 400 and 500 W; T_{air} – 35, 40 and 45 °C V_{air} – 1.0, 1.25 and 1.5 m/s.	t_{drying} reduction – 2.25 times D_{eff} – 0.21×10^{-10} – 1.57×10^{-10} m^2/s	Rehydration ratio 4.5–5.3	Sharma, Verma and Pathare, 2005a
	MIR (2.4 – 3.0 μm); T_{dry} – 60, 70 or 80 °C; slice thicknesses 2, 4 or 6 mm; T_{air} – 30, 40 or 50 °C; V_{air} – 0.8, 1.4 and 2.0 m/s; FMC – 7–8% (wb)	t_{drying} – 220 min (IR – air 40 °C); 280 min (IR); 340 min (hot air at 60 °C)	Pyruvic acid content 16.95 μmol/g (IR–air); 9.83 (IR); 10.96 (hot air) ΔE – 20.63 (IR–air) – 30.62 (IR); Browning index 13.74 (IR–air) – 18.99 (hot air)	Kumar *et al.*, 2005 Jain and Pathare, 2004 Pathare and Sharma, 2006
	IR intensity, 2.65, 3.53, 4.42 W/cm^2; T_{air} – 35, 40, 45 °C: V_{air} – 1.0, 1.25, 1.5m/s	Nine drying models were fitted	Asymptotic model fitted well r – 0.9995, standard error – 0.0064, χ^2 – 4.4×10^{-5} –	
	IR intensity –26.5 – 44.2 kW/m^2; T_{air} –30–60°C and V_{air} – 0 to 1.5 m/s; slice thickness 6 mm	D_{eff} – 0.2514×10^{-10} to 0.3233×10^{-10} m^2/s; $E_{activation}$ – 5.06 and 10.63 kJ/mol		
Carrot	IR power – 300, 400, 500 W; V_{air} – 1.0, 1.5, 2.0 m/s; slice – 29.6 dia and 6 mm thick; IMC – 8.52 to FMC – 0.11 kg/kg dm	t_{drying} – varied 252–277; 205–236; 145–155 min; E_{spe} – 12.22–14.58 MJ/kg water evaporated	ΔE – 10.08–15.59; shrinkage ratio – 0.160–0.230; Rehydration ratio – 2.72–4.78	Kocabiyik and Tezer, 2009
Carrot pomace (peel)	IR power levels – 83, 125, 167 and 209 W; IMC – 8.233; FMC – 0.015 kg/kg dm	t_{drying} – decreased from 310 to 60 min as IR power increased; D_{eff} – 0.59×10^{-10} – 3.40×10^{-10} m^2/s; $E_{activation}$ – 5.73 kW/kg	–	Doymaz, 2013

Sweet potato slices	IR power levels – 104, 125, 146 and 167 W using 250 W halogen lamp; slice thickness –3, 5, 8 mm; IMC – 81.8%(wb); FMC – 15%(wb)	Average drying rates increased 2.57 times as IR power increased from 104 to 167 W; t_{drying} – decreased from 270 to 105 min; D_{eff} – increased from 1.31×10^{-10} to 3.66×10^{-10} m^2/s	Maximum rehydration ratio was at 146 W	Doymaz, 2012a
Red pepper	IR power – 300, 400 and 500 W; V_{air} – 1.0, 1.5 and 2.0 m/s	t_{drying} – 314–455, 213–297 and 196–230 min; E_{spe} – 4.62–7.59 kW h/kg	ΔE – increased by 4.79–12.87; vitamin C decreased – 0.3085-1.382 mg/g compared to fresh (3.158 mg/g); shrinkage – 0.162–0.263; rehydration ratio – 2.10–3.78	Nasiroğlu and Kocabiyik, 2009
Apple slices	Nine IR lamps (175 W each) with peak wavelength 1200 nm; V_{air} – 0.5, 1.0 and 1.5 m/s; distance between emitters and drying surface – 10, 20 and 30 cm; apple slice – 5.5 mm	Influence of V_{air} on drying kinetics by IR was more pronounced than distance between IR and drying surface; water flux of 0.43–0.47 g/m^2 s at 10 cm distance of IR and surface; 50% reduction; t_{drying} compared to convective drying at 65 °C to 10% moisture	Shrinkage – four folds in thickness	Nowak and Lewicki, 2004

(*continued overleaf*)

Table 19.4 (*continued*)

Product	Energy attributes	Drying attributes	Quality attributes	References
Peach	Two-stage drying process; Far-IR drying (0.50, 0.70 and 1.00 kW/kg), followed by MW drying (0.50, 0.70 and 1.00 kW/kg); IMC – 9.36 kg/kg dm	Two falling rates – above 1.7(db) and another below 1.7(db); lower dehydration rate with IR at same power level except accelerated rate at initial period of drying; $E_{specific}$ inversely proportional to MW power	Sensory quality was highest at IR power 2.891 kW/kg and energy consumption rate was lower at 2.860 kW/kg	Wang and Sheng, 2006
Banana slices	Two modes of energy supply in drying – low-pressure superheated steam and far-IR drying (LPSSD–FIR) and vacuum–FIR drying Sequential IR and freeze-drying (SIRFD) at IR intensity of 3, 4 and 5 kW/m^2 and 62.5 °C used for hot-air drying; first pre-dehydrating of banana slices (5 mm thick) with IR followed by freeze drying	LPSSD–FIR took longer drying time than vacuum–FIR at all drying conditions except at the highest drying temperature tested (90 °C) IR heating took 3.1 and 6.2 min for 20 and 40% weight reduction with 5 kW/m^2; hot air took 11.2 and 37.3 min; corresponding FD took 23 and 38 h pre-dehydrated nonacid dipping and 21.1 h for non-pre-dehydrated slice	LPSSD–FIR at 80 °C gave best drying condition for crispy and less darker banana slices SIRFD produced high crispy banana chips and improved product colour (acid dipped); reduced freeze-drying time; pre-dehydrated nonacid dipped samples took longer time in freeze drying	Nimmol *et al.*, 2007, Pan *et al.*, 2008
Grape by-products	Two 200 W halogen lamps were used as source of infrared; T_{drying} was programmed (100, –160 °C); sample size – 25 g; sample thickness – 0.70 cm; IMC – 204.32%(db); FMC – 38.98%(db)	t_{drying} varied from 60.5 to 21 min for T_{drying} and moisture range; D_{eff} – 11.01×10^{-9} to 26.05×10^{-9} m^2/s; $E_{activation}$ – 19.27 kJ/mol; Midilli model was found suitable for prediction		Ruiz Celma *et al.*, 2009

Cashew nut	FIR ceramic heater (3–15 μm); product temperature – 100, 110 and 120 °C; heater to product distance 36 mm Quartz lamp (1 kW, peak wavelength 1.1–1.2 μm, 0.2 W/m^2); T_{air} – 55–95 °C	Fick's unsteady state law for spherical configuration was found to hold good; D_{eff} – 0.948×10^{-9} to 2.2×10^{-9} m^2/s; $E_{activation}$ – 28.7 kJ/mol t_{drying} – 15–55 min; best drying condition for removal of testa was 55 min at 55 °C	–	Hebbar and Rastogi, 2001, Hebbar and Ramesh, 2005
			Colour increased with increase in T_{drying} and t_{drying}, indicating darkening of kernel colour;. imparted desired brittleness in kernels	
Pomegranate seeds	Halogen lamp 83 to 146 W; seeds IMC 43.6% (wb)	t_{drying} reduced from 150 to 60 min with increase in IR power; good prediction models were Page, Midilli and Weibull; D_{eff} – 1.96×10^{-11} to 6.29×10^{-11} m^2/s; $E_{activation}$ – 10.72 kW/kg		Doymaz, 2012b

IR, infrared radiation; T_{air}, temperature of air; V_{air}, velocity of air; t_{drying}, drying time; D_{eff}, effective diffusivity; E_{spe}, specific energy consumption; $E_{activation}$, activation energy; ΔE, total colour difference; IMC, initial moisture content; FMC, final moisture content; NIR, near-infrared; MIR, medium-infrared; FIR, far-infrared; dm, dry matter.

19.3.2 Baking

Baking, a high temperature process (200–250 °C) for final heating of dough, proceeds under three modes of heat transfer (radiation, convection and conduction), and brings about several changes in physical, chemical and biochemical aspects. Along with essential maintenance of the humidity level inside the baking chamber, the rate and quantum of energy absorbed are important factors for final quality of the baked product (Therdthai, Zhou and Adamczak, 2002). Heating with various energy systems in baking are infrared, microwave, jet impingement and combined heating.

Biscuits and crackers Baking a wide range of biscuit products (crackers, semi-sweet and short dough types) on the usual type of baking support (wire mesh or a continuous steel band) using quartz–tungsten tubes emitting IR radiation with a peak wavelength of 0.2 μm showed a 50% reduction in baking time than in a conventional oven without significant difference in appearance, physical dimension and eating properties (Wade, 1987). Further, checking (spontaneous breakage) of Rich Tea biscuits baked by NIR at a peak wavelength of 1.2 μm could be completely avoided on storage, even at moisture contents that would expect 100% checking in conventionally baked products. This phenomenon could be associated with the smaller difference in moisture content between the biscuit centre and biscuit rim in freshly NIR-baked biscuits compared with conventionally baked products. The performance of infrared-based systems for baking rice crackers, evaluated in comparison to conventional ovens (Sakai and Hanzawa, 1994), showed that the baking time and cost for energy were 66.7 and 54.5% of that with a conventional heating oven, respectively.

Bread and cakes Baking with short-wave IR, with its high heat transfer efficiency, penetration capability and rapid control of oven parameters, makes it worthwhile in the bread-making process. This technique could attain desirable quality of bread in addition to reduced baking time (Skjöldebrand and Andersson, 1989). Typically, a 10 mm dough piece would take 6 min to obtain the desired crust colour in an IR oven compared to 16 min in conventional baking in an oven at 230 °C, and IR baked bread would produce a thinner crust and softer crumb. Optimum crust thickness could be obtained by lowering the power level at the second stage of baking. Radiant heating with an NIR heater led to a greater heat sink into the food samples, resulting in the formation of relatively wet crust layers, compared to dry layers formed by FIR heaters (Sakai and Hanzawa, 1994). However, the rate of colour development by FIR heaters was greater with NIR heaters, primarily due to a more rapid heating rate on the surface.

In baking cakes using far-infrared energy, the batter temperature increased faster than that in an electric oven (Shyu *et al.*, 2008). The hardness of sponge

cake baked in a far-infrared oven would remain softer even after 7 days storage than that of a sponge cake baked in an electric oven. No significant differences in quality parameters of baking products, such as volume, water activity, staling rate or sensory scores, were observed between these two baking techniques.

Tortillas Infrared baking of wheat flour tortilla for 17–19 s using a selected wavelength band and emission temperatures of 549 or 584 °C showed good characteristics of rollability, puffing, layering, colour and texture of the final product in addition to less consumption of energy (Martínez-Bustos *et al.*, 1999). The loss of moisture during baking of the tortillas formed by hot-pressing and baked by IR was significantly lower than that of tortillas baked by the traditional method of cooking on a hot griddle and commercial tortillas cooked in a three-tier, gas-fired oven (Martínez-Bustos *et al.*, 1999).

Combined heating modes of baking Baking with a microwave (MW) oven lacks characteristic brown colour development on the surface of the baked products. Several patents have been documented on the use of IR heating in combination with MW heating to impart surface browning in breads, biscuits and cookies (Jung and Lee, 1992; Fujii and Tsuda, 1987; Eck and Buck, 1980). A combination of microwaves with other modes of heating were also found to improve uniformity of microwave heating and provide more control on the moisture transport while increasing the speed of heating (Datta, Greedipalli and Almeida, 2005). Additionally, heat transfer mechanisms are qualitatively different in infrared and jet-impingement heating from that of microwave heating alone.

Combined modes of heating using IR and microwave heating could be optimally used for baking bread (Demirekler, Sumnu and Sahin, 2004). There would be no significant difference in the quality of the bread with baking in a halogen lamp–microwave combination oven optimally set with 70% upper halogen lamp power, 50% lower halogen lamp power and 20% microwave power. The baking time of bread could be reduced by 60% in a combined heating system than that with a conventional heating system.

In halogen lamp–microwave combination-baking, breads baked at 70% halogen lamp power and 30% microwave power for 3 min could give colour change (from dough) and a specific volume similar to conventionally baked breads (Keskin, Sumnu and Sahin, 2004). However, the firmness of breads was still higher as compared to conventionally baked ones. An extension of baking time resulted in darkening the colours with decreased specific volume and increased firmness to an unacceptably great extent.

Breads were baked with three different modes of heating oven, viz. microwave plus infrared (MIR), microwave plus jet impingement (MJET) and jet impingement (JET). The MIR oven gives bread with a significantly lower specific volume, porosity and moisture content than that with MJET

and JET ovens (Sumnu *et al.*, 2007). Breads baked in the JET oven had the highest total porosity followed by MJET and MIR (Datta *et al.*, 2007).

Cakes baked in an IR–microwave combined mode-heating oven could give similar colour and firmness compared to those with the conventional baking process using an electric oven except there was low specific volume and high weight loss with the former system (Sumnu, Sahin and Sevimli, 2005). The best conditions for baking in an IR–microwave oven could be a combination of 70% halogen lamp power and 50% microwave power, which would reduce the baking time by as much as 75%. The IR–microwave combination heating mode produced breads with similar staling degrees as conventionally baked ones (Ozkoc *et al.*, 2009).

Nevertheless, it is not uncommon to have undesirable effects on the quality of baked products with IR energy assisted baking. Baking of breads using only the halogen lamp mode of the combined halogen–MW oven would form a very thick bread crust (Keskin, Sumnu and Sahin, 2004). Cakes, baked in an IR-microwave combination oven, gave low specific volumes and higher weight loss (Sumnu, Sahin and Sevimli, 2005); Cookies made with a convection oven showed good sensory qualities, such as flavour, texture and surface browning, better than with a conventional deck or IR ovens (Heist and Cremer, 1990). However, energy used in baking was highest in the convection oven.

Effect of ingredients on baking The effects of different gums, viz. xanthan, guar, xanthan–guar blend and κ-carrageenan, on the quality of breads baked with an infrared–microwave combination oven have been studied. Undesirable final bread quality with 0.5% κ-carrageenan in the bread formulation was observed while addition of the same level of xanthan–guar improved bread quality, such as high specific volume and porosity and low hardness values (Keskin, Sumnu and Sahin, 2007). Addition of xanthan–guar gum in the blend (0.5%) decreased hardness, retrogradation enthalpy and total mass crystallinity values of bread samples, suggesting that the staling process could be delayed (Ozkoc *et al.*, 2009). Colour and hardness for both types of baking schemes were found to be dependent on the formulation.

Cakes containing a fat replacer consisting mostly of whey protein, baked in microwave and near-infrared–microwave combination ovens were found to be the firmest cakes (Sakiyan *et al.*, 2007).

The degree of starch gelatinization in baked cakes was found to be insufficient (55 to 78%) in a microwave oven (depending on the formulation); contrary to that, there was a 85 to 93% gelatinization in a conventional oven (Sakiyan *et al.*, 2011). Combining infrared with microwaves increases it to 70–90%. Further, addition of fat enhanced the starch gelatinization process in microwave and infrared–microwave combination ovens. However, under such conditions, reduction in starch gelatinization was observed in a conventional baking oven.

19.3.3 Roasting

The roasting process enhances several sensory qualities of nuts and seeds. It proceeds with volatilization of components like free amino acids, peptides, fatty acids, vitamin E along with some chemical changes and moisture loss. The mechanisms leading to defined mixtures of free amino acids and peptides, and involvement of precursor proteins during roasting of coffee beans were studied (Montavon, Mauron and Duruz, 2003). The increases of the sterols and vitamin E during the roasting process of pumpkin seeds could be attributed to the changes of the meal, since at the end of roasting the oil emerges from the seeds, resulting in altered chemical behaviour of the extraction process (Murkovic *et al.*, 2004). In conventional roasting, the material is heated to between 130 and 150 °C for 40 to 45 min (depending on the extent of roasting). Excessive heating at high temperatures adversely impairs the aroma profile.

Nuts and oilseeds IR assisted roasting started as early as the late 1960s (Arnold, 1968) using an infrared tunnel for roasting green pecan nutmeats coated with vegetable gum (to retard rancidity), salt and spices on an open mesh type moving belt under low ambient heat with a final moisture content of 0.3–0.8%. Whole peanut kernels roasted by infrared took less roasting time and lost more oil even though the moisture content was fairly high after roasting (Mahajan and Pai, 1988). Roasting of whole peanut kernels in a conventional oven at 150 °C took a long time (15 min) to remove 62.59% of the total oil in it, while the final moisture went as low as 1.74%. IR roasting at 230 °C took only 3 min to extract 75.30% of the total oil with a final moisture content of 3.52%. Good oil recovery with very good quality of defatted kernels might be attributed to sufficient cell disruption without excess moisture loss in the IR roasting.

Microwave–infrared combination oven roasting for hazelnuts revealed that the roasting time could be reduced by 87.5% at optimum conditions with 90% microwave power, 60% upper halogen lamp power and 20% lower halogen lamp power compared to a conventional electric oven at 150 °C and 20 min (Uysal, Sumnu and Sahin, 2009). Further, hazelnuts roasted at this optimum point had comparable quality with conventionally roasted ones with respect to colour, texture, moisture content and fatty acid composition.

Almonds were roasted at 130, 140 and 150 °C with infrared (IR), sequential infrared and hot air (SIRHA) and traditional hot air (HA). The results showed 4.10, 5.82 and 6.96 log bacterial reductions in terms of the organism *Pediococcus* sp. NRRL B-2354 (as a surrogate for *Salmonella enteric Enteriditis* PT 30) with 38, 39 and 62% saving in time compared to HA roasting (Yang *et al.*, 2010). The sensory quality of medium roasted almonds showed no significant difference between these roasting methods. It was concluded that the SIRHA roasting would be a promising method for the production of dry-roasted pasteurized almonds. In their subsequent study, the shelf-life

of roasted almonds with similar roasting processes followed by packaging in paper bags and storing at 37 °C revealed no change in colour, peroxide value, moisture content, water activity, volatile components and sensory quality during the first three months of storage (Yang *et al.*, 2013). However, concentration of aliphatic aldehydes in the nuts differed significantly with the method of roasting and increased significantly in all roasted samples during storage.

IR (near-infrared, 1.1 to 1.3 μm, 6 kW) roasting of sesame seeds at 200 °C for 30 min increased the efficiency of conversion of lignin sesamolin to sesamol (51 to 82%) compared to conventional drum roasting, along with increased oxidative stability of sesame oil synergistically with tocopherols (Kumar, Rao and Singh, 2009). The functional properties of defatted flours obtained from either IR roasted or conventionally roasted sesame seeds remained the same.

Green tea The aroma that is special to the fired tea is thought to be created when the leaves are dried and heated to between 100 and 120 °C. Bitterness and astringency in lower-grade tea leaves could be eliminated by toasting it at around 200 °C (torrefaction process), which breaks down polyphenols into pyrrole and pyrazine types of flavour components (Sakai and Mao, 2006). The hot-air heating method has been substituted with FIR (or microwaves and FIR in combination) heating to heat the tea leaves more uniformly, and produces tea with a better flavour. FIR roasting and drying (90 °C for 10 min) resulted in the manufacture of high-quality green tea (Kim, Jeong and Jo, 2006) with a significant increase in total polyphenols, flavanols, epigallocatechin and epigallocatechin gallate contents to 811.1, 208.7, 89.88 and 16.33 mg/g, respectively, from corresponding values of 475.6, 175.7, 57.68 and 9.60 mg/g with the control samples. Ascorbic acid, caffeine and nitrite scavenging activities also increased with negligible change in the overall colour.

Coffee beans A patented roasting unit for coffee beans using infrared heating has been developed (Ito *et al.*, 1989). The rotating body comprised a horizontal rotary drum. The drum stirred the coffee beans. Infrared heaters disposed on the inner wall surface of the roasting body surrounded the rotary drum in order to heat the coffee beans accommodated in the rotary drum by radiant heat and a circulating path circulated the hot air in the rotary drum into the roasting body in order to utilize the thermal energy effectively.

An oven for FIR roasting of coffee beans has been developed as a substitute for the hot-air roasting system (usually 350–450 °C). This oven consisted of an array of ceramic heaters, installed over a perforated rotating drum. The beans inside the drum were irradiated. A rapid increase in the temperature of the beans with FIR radiation occurred that accelerated the chemical reaction leading to roasting (Sakai and Mao, 2006). In the FIR roasting process, the formation of chlorogenic acid, which would be important in respect to taste, was almost the same for both roasting methods, while smaller quantities of other

organic acids were produced in FIR roasting than those in hot-blast roasting. Thus, FIR roasted coffee beans seemed to have a delightful and mild taste.

Grains A fuzzy rule-based control system for an electric infrared roaster of cereal grain was developed for wheat (Brown, Rothwell and Davidson, 2001). The bulk average temperature of the grain exiting the roaster was used to adjust the infrared panel temperature and ensured an adequate degree of roast. The control system was tested in-plant while roasting cracked wheat.

Roasting of other foods Roasting fish pastes using FIR heating showed an increase in the production rate of 294% and a reduction in energy consumption of 26.1% compared to that of an electric oven (Sakai and Hanzawa, 1994). Sweet potato or chestnut becomes sweeter and more delicious when roasted with FIR. Since, FIR energy permeates into the food, it uniformly heats the potato inside (Sakai and Mao, 2006).

Roasting to increase antioxidant activity An increase in the antioxidant activity of peanut (*Arachis hypogaea* L.) hull was observed after it was FIR-irradiated or heat-treated at 150 °C for 5, 10, 15, 20, 40 or 60 min (Lee *et al.*, 2006). The total phenol content (TPC), radical scavenging activity (RSA) and the reducing power of water extract (300 mg/10 ml) of peanut hull (WEPH) increased from 72.9 to 141.6 μM, 2.34 to 48.83% and 0.473 to 0.910, respectively, compared to the untreated controls after it was treated with far-infrared (FIR) radiation. Heat-treated peanut hulls at 150 °C for 60 min showed these changes to be much lower than those of the FIR-treated samples. FIR radiation on to rice hull could liberate and activate covalently bound phenolic compounds that have antioxidant activities (Lee *et al.*, 2003). Radical scavenging activity and total phenol content of rice hull extracts increased from 47.74 to 79.63% and from 0.12 to 0.19 mM, respectively, after 30 min of FIR roasting of rice hulls, while simple heat treatments could not cleave covalently bound phenolic compounds.

19.3.4 Blanching

This is an important step in canning, freezing or drying processes for fruits and vegetables. It facilitates inactivation of oxidative enzymes, fixation of green colour of chlorophylls, removal of trapped air in the mass and modification of texture for proper fill-volume in canning and enhancement of internal moisture diffusion in drying. It is carried out by heating the material in hot water (80–90 °C for 5–8 min) or exposing it to steam for a short duration.

Mechanism of the IR blanching process The mechanism of blanching of fruits and vegetables by IR radiations follows absorption of energy selectively

by water molecules present in it. Specific spectral bands of IR radiation (medium- and far-IR) with peak wavelengths at 3, 4.7 and 6 μm penetrate into the product; the energy is absorbed by the water molecules (moisture) and subsequently increases the temperature of the mass (Ginzberg, 1969). From a heat and mass transfer viewpoint, IR assisted blanching is a complex phenomenon and needs a proper modelling to adopt control of the process.

IR blanching of vegetables No quality differences occurred for blanching of leafy vegetables (endive and spinach) using infrared and radio frequency treatments compared to conventional hot water and steam blanching. However, blanching using microwave energy alone or in combination with steam showed an improvement in vitamin C retention and better sensory characteristics (Ponne, Baysal and Yuksel, 1994). Mild heat treatment of carrot slices as a pre-treatment for frozen storage was accomplished (Galindo, Toledo and Sjöholm, 2005). FIR heating (810 K, peak wavelength 5 μm) of carrot slices (3.0–3.2 diameter and 0.7 cm thick) for 7 s showed damaged cells only in the first half millimetre from the surface and preserved most of the texture characteristics of the raw tissue compared to blanching the same in boiling water (5 to 30 s). No viable cell in carrot slice could be identified with blanching in boiling water for 30 s.

Simultaneous infrared dry blanching and dehydration (SIRDBD) This is a new hybrid one-step process of heating for blanching and partial drying of fruits and vegetables (Pan and McHugh, 2006), and finally gives a high product quality. This combined one-step process claims to have higher efficiency than the conventional two-step process. The basic mechanism involved here was to use catalytic infrared (CIR) radiation that facilitated inactivation of enzymes responsible for oxidative deterioration of fruits and vegetables. A specific CIR emitter generated a peak wave length of radiation (between 3 and 6 μm) that matched the absorption peak wavelength of water.

Two modes of SIRDBD could be adopted – continuous and intermittent heating. Continuous heating was advantageous in relatively fast delivery of high energy to the surface of the product, quick heating-up and rapid moisture removal or enzyme inactivation. Processing and quality characteristics of apple slices under SIRDBD with continuous heating (Zhu and Pan, 2009) showed high radiation intensity. Thin slices of apple had a faster increase of product temperature, quicker moisture removal and inactivation of polyphenol oxidase (PPO) and peroxidase (POD). Thus 90% inactivation of POD in apple slices could be achieved with thicknesses from 5 to 13 mm in 2–15 min. The resulting moisture reductions were in the range of 15–49% while the surface colour of the apple slices were preserved well (Zhu and Pan, 2009).

The problem of limited penetration of far-infrared (FIR) and the application of FIR on thick materials could be solved with intermittent heating (Sandu, 1986). In addition, energy savings and improved product quality are

additional advantages of intermittent heating since the desired processing temperature could be maintained (Chua and Chou, 2003). The best processing parameters for the intermittent heating mode for apple slices are a surface temperature of 75 °C, 5 mm slice and 7.5 min processing time, which lead to a final product with 10% residual POD, less than 1% residual PPO, 36% moisture reduction and an overall surface colour change of 2.27 (Zhu *et al.*, 2010).

The developed heat and mass transfer model for apple slices (5, 9 and 13 mm thick) under the SIDBD process (4 kW/m^2, 10 min) indicated that it can predict temperature, moisture profiles and inactivation rate of enzymes (PPO and POD), vis-à-vis blanching and dehydration performance (Lin *et al.*, 2009).

Effects of blanching on enzymes Reduction in the Michaelis and Menton constant (K_m) and maximum velocity of the specific reaction (V_{max}) with a model enzyme, such as xanthine oxidase, was found to be 51 and 85% of the non-irradiated control, respectively, compared to heat inactivation with an FIR radiation energy of a 5 μm wavelength and flux density of 72 mW/cm^2 (Kohashi, Akao and Watanabe, 1993). Inactivation of lipase and α-amylase by far-infrared (FIR) radiative heating and heating by thermal conduction was found to be mostly similar as long as the temperature profiles of the enzyme solutions were identical (Sawai *et al.*, 2003). Some enzymes are responsible for development of off-flavour in peas. FIR could be successfully used to inactivate these enzymes prior to freezing the peas (van Zuilichem, Vant Reit and Stolp, 1986),

19.3.5 Pasteurization and sterilization

Infrared heating could be used for pasteurization and sterilization with high thermal efficiency and a fast heating rate compared to a conventional heating process using steam (Krishnamurthy *et al.*, 2008a). The underlying mechanism of killing microorganisms is that absorption of IR energy by water molecules in the cell mass (cytoplasmic fluid) precedes a rapid rise in its temperature, and consequently increases thermal death rates of all types of microorganisms. This includes vegetative cells and spores of bacteria, yeast and molds. The food could be either liquid or solid.

The infrared power level, temperature of the food, peak wavelength and bandwidth of the infrared emitter, sample depth, nature and level of microorganisms, moisture content, state of the growth phase of the microorganism (exponential or stationary phase) and types of food materials are dominant factors associated with the efficacy of microbial inactivation by infrared heating.

Surface infrared pasteurization of fresh cottage cheese (packed in plastic containers) at 71 °C for 5 min reduced the number of contaminating yeast and molds in a surface layer of about 1 cm deep by at least 1 order of magnitude

and improved the shelf-life of approximately 3–4 weeks when stored at 4 °C (Rosenthal, Rosen and Berstein, 1996).

At the given temperatures, the death rate constant (k_{death}) of the test organism *E. coli* with FIR heating was larger than k_{death} values with conductive heating, suggesting that FIR heating may allow a given pasteurization target level to be achieved at lower temperatures than by conductive heating, while maintaining enzyme activity levels (Sawai *et al.*, 2003).

IR heating is also used to decontaminate food-contact surfaces to eliminate microorganisms and to enhance the shelf-life of food products, for example heating baking trays before dough was put on them. Concise information about the application of IR radiation for pasteurization and sterilization of different foods is presented in Table 19.5.

Selective infrared radiation (IR) heating for the inactivation of fungal spores based on a dynamic temperature profile was explored (Jun and Irudayaraj, 2003) with an integrated model that combined thermal death kinetics and heat transfer. Selective IR heating was found to differentially contribute to a higher degree of lethality compared to normal IR heating. Denaturation of protein components in the selective IR range also contributed to an additional increase in the degree of lethality of fungal spores, compared to the model prediction.

Combined FIR heating was carried out at 150, 200 and 250 °C and forced airflow of 0.2 m/s at 20 °C for surface decontamination of strawberry (Tanaka *et al.*, 2007). Monte Carlo FIR radiation simulations (both internal heat transfer by conduction and external heat transfer by convection and surface radiation) were performed in the CFD code ANSYS CFX5.7 to investigate the suitability of the method. More uniform surface heating of the fruits could be achieved with FIR heating than with air convection heating, and the maximum fruit temperature was 50 °C; it was well below the critical limit at the same average temperature. A better configuration consisted of FIR heaters on four sides combined with a cyclic heating operation to avoid fruit damage. However, the method was found to be inferior to water bath heating in terms of efficiency and uniformity.

19.4 Conclusions

IR radiation heating is an environmental friendly technology. It could be applied conveniently to different food processing systems with several advantages over the corresponding conventional methods. The list of foods that could be treated with IR heating includes fruits, vegetables, spices, juices, milk, nuts, etc; the list is also increasing. The limitation of surface heating with IR could be widened to bulk heating by a combination of IR heating with convective, conductive and radiation (MW, radio frequency, UV, etc.) modes of heating. In order to achieve high efficiency in the IR heating process,

Table 19.5 Application of IR radiation for pasteurization and sterilization of foods

Food Item	Microorganism	Energy attributes	Effect	Reference
Wheat	*Bacillus subtilis*	IR power levels of 0.5, 1.0, 1.5 and 2.0 kW; surface temperature of 45, 65, 95 and 120°C	$\log_{10}$ CFU/g total bacteria 0.83, 1.14, 1.18 and 1.90, respectively after 60 s of heating	Hamanaka *et al.*, 2000
Fig fruits	*Rhodotorula mucilaginosa*	Sequential treatment with UV radiation and IR heating carried out; peak wavelength of the IR heater and UV lamp were 950 and 253.7 nm, respectively, with corresponding energy density of 14.8 and 31.7 mW/cm^2	Effective in the surface decontamination of fig fruits from yeast and fungus occurred with IR heating for 30 s followed by UV irradiation for 30 s	Hamanaka *et al.*, 2011
Cottage cheese	Yeast and molds	Surface heating with infrared lamp of 250 W at a distance of 2.5 to 3 cm from surface	Effective surface pasteurization was achieved with initial count of < 10 cells/g and $\log_{10}$ reduction of < 3 cells/g treated sample after 8 weeks of storage	Rosenthal, Rosen and Berstein, 1996
Milk	*Staphylococcus aureus*	IR lamp temperatures – 536 and 619 °C; volumes of samples – 3, 5 and 7 ml with treatment times of 1, 2 and 4 min	Complete inactivation occurred in 4 min at 619 °C (8.41 $\log_{10}$ CFU/ml); more than 5 min at both 536 and 619 °C resulted in no detectable colonies	Krishnamurthy *et al.*, 2008a
Hot dogs	*Listeria monocytogenes*	Quartz tube infrared heating element (1250 W); average initial inoculum of 7.32 log (CFU/g) taken	Surface pasteurization was effective in activating *L. monocytogenes* in RTE meats; 3 min holding at 80 °C or 2 min at 85 °C; a total of 6.4 or 6.7 log of *L. monocytogenes* were inactivated	Huang and Sites, 2008
Almond	*Salmonell enteritidis*	IR power 5500 W/m^2 and time of exposure 30, 35, and 45 s corresponding to temperatures of 90, 102 and 113 °C	Log reductions were 0.63, 1.03 and 1.51 corresponding to stated exposure time and temperature; kernel colour was indistinguishable from those of untreated kernels	Maria *et al.*, 2008

(*continued overleaf*)

Table 19.5 (*continued*)

Food Item	Microorganism	Energy attributes	Effect	Reference
Nonspecific	*S. aureus and E. coli*	The radiative power irradiated on the bacterial suspension was 3.22 kW/m^2; decreasing the sample depth accelerated inactivation of spores	Reduction in microbial population at 321 K *S. aureus-2* and 5 $\log_{10}$ CFU/ml corresponding to sample depths of 2.9 and 0.9 mm *E. coli* – 1.33 and 1.66 $\log_{10}$ CFU/ml corresponding to 1.3 and 2.2 mm	Hashimoto *et al.*, 1992
	Bacillus subtilis	IR heaters having peak wavelengths of 950 (heater A), 1100 (heater B) and 1150 nm (heater C) with corresponding energy flux of 4.2, 3.7 and 3.2 μW/cm^2 nm	Pathogen inactivation was higher with heater A than those of heaters B and C. Spores at approximately 0.9, 0.7 and 0.6 a_w were most resistant to IR heating at wavelengths of 950, 1100 and 1150 nm, respectively	Hamanaka *et al.*, 2006
	E. coli	FIR heater of wavelength 4 to 7 μm	Reduction in microbial population after 2 min of exposure was 0.76, 0.90 and 0.98 log-cycles at 56, 58 and 60°C, respectively	Sawai *et al.*, 2003
Corn meal	*Aspergillus niger and Fusarium proliferatum*	IR radiation heating with selective spectral band of 5.88 to 6.66 μm	40% increase in inactivation of *A. niger* and *F. proliferatum* compared to nonspecific IR radiation heating	Jun and Irudayaraj, 2004
Paprika powder	*Bacillus cereus* spores	Powder with water activities (a_w) of 0.5, 0.8 or 0.96 was exposed to NIR at 11 and 25 kW/m^2 (1.2 μm) and MIR at 5 and 11 kW/m^2(2.7 μm) NIR at 1.2 μm (2100 °C); a_w values of 0.84 and 0.88; pH of 4.0 and 4.5	At a_w 0.5 and 0.8 resulted in unsatisfactory spore reduction; at a_w 0.96, poor reduction occurred at the surface but complete inactivation; at a detection limit of 1 $\log_{10}$ CFU/g occurred in areas with high a_w Increase in temperature from 90 to 95 °C showed higher inactivation than 95 to 100 °C; powder with a_w of 0.88 and heated to 95–100 °C reduced *B. cereus* spore concentration of 4.5 $\log_{10}$ CFU/g within 6 min; lowering pH to 4.0, spore reduction was not significantly higher than at pH 4.5	Staack *et al.*, 2008a Staack *et al.*, 2008b

emitters with a peak radiation band matching the absorption band of water in food should be employed. Reasonable control for radiation intensity, exposure time and agitation of materials can moderate the damage and preserve the quality of the products to a large extent. IR heating is a complex phenomenon in achieving inactivation of different groups of microorganisms. A proper model simulating simultaneous kinetics in the combined mode of heating is highly desirable. Successful commercial applications of IR heating in the food processing sector with sustained benefits in cost, productivity and quality of finished product still remains a challenge, and demands extended research.

References

Afzal, T.M., Abe, T. and Hikida, Y. (1999) Energy and quality aspects during combined FIR convection drying of barley. *Journal of Food Engineering*, **42**, 177–182.

Almedia, M., Torrance, K.E. and Datta, A.K. (2006) Measurement of optical properties of foods in near and mid infrared radiation. *International Journal of Food Properties*, **9**, 651–654.

Arnold, F.W. (1968) Infrared roasting of coated nutmeats. US Patent **3**, 383–220.

Bal, S., Wratten, F.T., Chesnen, J.L. and Faulkner, M.D. (1970) An analytical and experimental study of radiant heating of rice grain. *Transactions of American Society of Agricultural Engineers*, **13**(5), 644–652.

Brown, R.B., Rothwell, T.M. and Davidson, V.J. (2001) A fuzzy controller for infrared roasting of cereal grain. *Canadian Biosystems Engineering*, **43**, 3.9–3.15.

Chua, K.J. and Chou, S.K. (2003) Low cost drying methods for developing countries. *Trends in Food Science and Technology*, **14**, 519–528.

Chua, K.J., Chou, S.K., Mujumdar, A.S., Ho, J.C. and Hon, C.K. (2004) Radiant–convective drying of osmotic treated agro-products: effects on drying kinetics and product quality. *Food Control*, **15**, 145–158.

Dagerskog, M. and Osterstrom, L. (1979) Infra-red radiation for food processing. I. A study of fundamental properties of infrared radiation. *Lebensmittel Wissenschaft und Technologie*, **12**, 237–242.

Das, I., Bal, S. and Das, S.K. (2003) Optimization of the vibration-aided infrared heating process using response surface methodology: an application to the drying and parboiling of high moisture paddy. ASAE Annual Meeting, Las Vegas, USA, Paper No. 036040.

Das, I. and Das, S.K. (2010) Emitters and infrared heating system design. In: *Infrared Heating for Food and Agricultural Processing*, Pan, Z. and Atungulu, G.G. (eds). CRC Press, Boca Raton, Florida, USA, pp. 57–88.

Das, I., Das, S.K. and Bal, S. (2004) A study of the drying performance of a batch type vibration aided infrared (IR) dryer. *Journal of Food Engineering*, **64**, 129–133.

Datta, A.K. and Almeida, M. (2005) Properties relevant to infrared heating of foods. In: *Engineering Properties of Foods*, Rao, M.A., Rizvi, S.S.H. and Datta, A.K. (eds). CRC Press, Boca Raton, Florida, USA, pp. 209–235.

Datta, A.K., Greedipalli, S.S.R. and Almeida, M. (2005) Microwave combination heating. *Food Technology*, **59**, 36–40.

Datta, A. K., Sahin, S., Sumnu, G. and Keskin, S.O. (2007) Porous media characterization of breads baked using novel heating modes. *Journal of Food Engineering*, **79**, 106–116.

Decareau, R.V. (1985) *Microwaves in the Food Processing Industry*, Mudgett, R.E. (ed.). Academic Press, Orlando, Florida, USA.

Demirekler, P., Sumnu, G. and Sahin, S. (2004) Optimization of bread baking in halogen lamp–microwave combination oven by response surface methodology. *European Food Research and Technology*, **219**, 341–347.

Dostie, M., Seguin, J.N., Maure, D., Ton-That, Q.A. and Chatingy, R. (1989) Preliminary measurements on the drying of thick porous materials by combinations of intermittent infrared and continuous convection heating. In:, *Drying '89*, Mujumdar, A.S. and Roques, M.A. (eds). Hemisphere Publications, New York, USA.

Doymaz, I. (2012a) Infrared drying of sweet potato (*Ipomoea batatas* L.) slices. *Journal of Food Science and Technology*, **49**, 760–766.

Doymaz, I. (2012b) Drying of pomegranate seeds using infrared radiation. *Food Science and Biotechnology*, **21**, 1269–1275.

Doymaz, I. (2013) Determination of infrared drying characteristics and modelling of drying behaviour of carrot pomace. *Journal of Agricultural Sciences*, **19**, 44–53.

Eck, P. and Buck, R.G. (1980) Method of browning food in a microwave oven. US Patent **4**, 396–817.

Fasina, O.O, Tyler, R.T., Pickard, M., Zheng, G.H. and Wang, N. (2001) Effect of infrared heating on the properties of legume seeds. *International Journal of Food Science and Technology*, **36**, 79–90.

Fujii, M. and Tsuda, T. (1987) Microwave heating and infrared ray heating appliance. US Patent **4**, 803–324.

Galindo, F.G., Toledo, R.T. and Sjöholm, I. (2005) Tissue damage in heated carrot slices. Comparing mild hot water blanching and infrared heating. *Journal of Food Engineering*, **67**, 381–385.

Ginzberg, A.S. (1969) *Application of Infrared Radiation in Food Processing*. Leonard Hill Books, London, England.

Hall, C.W. (1962) Theory of infrared drying. *Transactions of the American Society of Agricultural Engineers*, **5**, 14–16.

Hamanaka, D., Dokan, S., Yasunaga, E., Kuroki, S., Uchino, T. and Akimoto, K. (2000) The sterilization effects of infrared ray on the agricultural products spoilage microorganisms (Part I), Paper No. 006090. ASAE Annual Meeting, Wisconsin, USA, July.

Hamanaka, D., Uchino, T., Furuse, N., Han, W. and Tanaka, S. (2006) Effect of the wavelength of infrared heaters on the inactivation of bacterial spores at various water activities. *International Journal of Food Microbiology*, **108**, 281–285.

Hamanaka, D., Norimura, N., Baba, N. Mano, K., Kakiuchi, M., Tanaka, F. and Uchino, T. (2011) Surface decontamination of fig fruit by combination of infrared radiation heating with ultraviolet irradiation. *Food Control*, **22**, 375–380.

Hashimoto, A., Sawai, J., Igarashi, H. and Shimizu, M. (1992) Effect of far-infrared irradiation on pasteurization of bacteria suspended in liquid medium below lethal temperature. *Journal of Chemical Engineering Japan*, **25**, 275–281.

Headly, V.E. and Hall, C.W. (1963) Drying of shelled corn vibrated in an infrared source. *Transactions of the American Society of Agricultural Engineers*, **6**, 148–150.

Hebbar, U.H. and Ramesh, M.N. (2005) Optimisation of processing conditions for infrared drying of cashew kernels with testa. *Journal of the Science of Food and Agriculture*, **85**, 865–871.

Heist, J. and Cremer, L.M. (1990) Sensory quality and energy use for baking of molasses cookies prepared with bleached and unbleached flour and baked in infrared, forced air convection and conventional deck ovens. *Journal of Food Science*, **55**, 1095–1101.

Henry, C.J.K and Chapman, C. (2000) *The Nutrition Handbook for Food Processor*. Woodland Publishing Limited, Cambridge, England

Huang, L. and Sites, J. (2008) Elimination of *Listeria monocytogenes* on hotdogs by infrared surface treatment. *Journal of Food Science*, **73**, M 27–31.

Il'yasov, S.G. and Krasnikov, V.V. (1991) *Physical Principles of Infrared Irradiation of Foodstuffs*. Hemisphere Publishing House, New York, USA.

Ito, T., Kino, T., Nakamur, A. and Tamaki, Y. (1989) Coffee beans roasting device. US Patent 4,860,461 A. http://www.google.co.in/patents/US4860461.

Jain, D. and Pathare, P.B. (2004) Selection and evaluation of thin layer drying models for infrared radiative and convective drying of onion slices. *Biosystems Engineering*, **89**(3), 289–296.

Jun, S. and Irudayaraj, J. (2003) Selective infrared heating system design and evaluation. *Drying Technology*, **21**(1), 51–67.

Jun, S. and Irudayaraj, J. (2004) An integrated dynamic model for fungal spore inactivation by selective infrared heating *Applied Engineering in Agriculture*, **20**, 481–485.

Jung, K.H. and Lee, S.Y. (1992) Microwave ovens with infrared rays heating units. US Patent **5**, 310–979.

Keskin, S.O., Sumnu, G. and Sahin, S. (2004) Bread baking in halogen lamp-microwave combination oven. *Food Research International*, **37**, 489–495.

Keskin, S. O., Sumnu, G. and Sahin, S. (2007) A study on the effects of different gums on dielectric properties and quality of breads baked in infrared-microwave combination oven. *European Food Research and Technology*, **224**, 329–334.

Kim, S.Y., Jeong, S.M. and Jo, S.C. (2006) Application of far-infrared radiation in manufacturing of process green tea. *Journal of Agricultural and Food Chemistry*, **54**, 9943–9947.

Kocabiyik, H. and Tezer, D. (2009) Drying of carrot slices using infrared radiation. *International Journal of Food Science and Technology*, **44**, 953–959.

Kohashi, M., Akao, K. and Watanabe, T. (1993) Non-thermal effect of ceramics radiation on folate structure and on xanthin oxidase inhibition by folate. *Journal of Nutritional Science Vitaminol*, **39**, 425–435.

Krishnamurthy, K., Jun, S., Irudayaraj, J. and Demirci, A. (2008a) Efficacy of infrared heat treatment for inactivation of *Staphylococcus aureus* in milk. *Journal of Food Process Engineering*, **31**, 798–816.

Krishnamurthy, K., Khurana, H.K., Jun, S., Irudayaraj, J. and Demirci, A. (2008b) Infrared heating in food processing: an overview. *Comprehensive Reviews in Food Science and Food Safety*, **7**, 2–13.

Kumar, C.M., Rao, A.G. and Singh, S.A. (2009) Effect of infrared heating on the formation of sesamol and quality of defatted flours from *Sesamum indicum* L. *Journal of Food Science*, **74**, 105–111.

Kumar, D.G.P., Hebber, H.U., Sukumar, D. and Ramesh, M.N. (2005) Infrared and hot-air drying of onions. *Journal of Food Processing and Preservation*, **29**, 132–150.

Lin, Y.P., Tsen, J.H. and King, V.A.E. (2005) Effects of far-infrared radiation on the freeze drying of sweet potato. *Journal of Food Engineering*, **68**, 249–255.

Lin, Y.L., Li, S.J., Zhu, Y., Binglo, G., Pan, Z. and McHugh, T.H. (2009) Heat and mass transfer modeling of apple slices under simultaneous infrared dry blanching and dehydration process. *Drying Technology*, **27**, 1051–1059.

Lee, S.C., Kim, J.H., Jeong, S.M., Kim, D.R., Ha, J.U., Nam, K.C. and Ahn, D.U. (2003) Effect of far infrared radiation on the antioxidant activity of rice hulls. *Journal of Agricultural and Food Chemistry*, **51**, 4400–4403.

Lee, S.C., Jeong, S.M., Kim, S.Y., Park, H.R., Nam, K.C. and Ahn, D.U. (2006) Effect of far-infrared radiation and heat treatment on the antioxidant activity of water extracts from peanut hulls. *Food Chemistry*, **94**, 489–493.

Mahajan, G.N. and Pai, J.S. (1988) Defatting of whole peanut kernels after infrared heating. *Food Chemistry*, **27**, 237–240.

Maria, B.T., Pan, Z., Steven, H., Zhu, Y. and Tara, M. (2008) Reduction of *Salmonella enteritidis* population sizes on almond kernels with infrared heat. *Journal of Food Protection*, **71**, 897–902.

Martínez-Bustos, F., Morales, S.E., Chang, Y.K., Herrera-Gómez, A., Martinez, M.J.L., Baňos, L., Rodriguez, M.E. and Flores, M.H.E. (1999) Effect of infrared baking on wheat flour tortilla characteristics. *Cereal Chemistry*, **76**, 491–495.

Modest, M.F. (1993) *Radiative Heat Transfer*. McGraw-Hill International Editions, New York, USA.

Montavon, P., Mauron, A.F. and Duruz, E. (2003) Changes in green coffee protein profiles during roasting. *Journal of Agriculture and Food Chemistry*, **51**, 2335–2343.

Murkovic, M., Piironen, V., Lampi, A.M., Kraushofer, T. and Sontag, G. (2004) Changes in chemical composition of pumpkin seeds during the roasting process for production of pumpkin seed oil. Part 1, Non-volatile compounds. *Food Chemistry*, **84**, 359–365.

Nasiroğlu, S. and Kocabiyik, H. (2009) Thin-layer infrared radiation drying of red pepper slices. *Journal of Food Process Engineering*, **32**, 1–16.

Nimmol, C., Devahastin, S., Swasdisevi, T. and Soponronnarit, S. (2007) Drying of banana slices using combined low-pressure superheated. *Journal of Food Engineering*, **81**, 624–633.

Nindo, C.I., Kudo, Y. and Bekki, E. (1995) Test model for studying sun drying of rough rice using far-infrared radiation. *Drying Technology*, **13**(1 and 2), 225–238.

Nindo, C. and Mwithiga, G. (2010) Infrared drying. In: *Infrared Heating for Food and Agricultural Processing*, Pan, Z. and Atungulu, G.G. (eds). CRC Press, Taylor & Francis, USA, pp. 89–99. ISBN: 978-1-4200-9099-4.

Nowak, D. and Lewicki, P.P. (2004) Infrared drying of apple slices. *Innovative Food Science and Emerging Technologies*, **5**, 353–360.

Ozkoc, S.O., Sumnu, G., Sahin, S. and Turabi, E. (2009) Investigation of physicochemical properties of breads baked in microwave and infrared-microwave combination ovens during storage. *European Food Research and Technology*, **228**, 883–893.

Pan, Z. and Atungulu, G.G. (eds) (2010) *Infrared Heating for Food and Agricultural Processing*. CRC Press, Taylor & Francis Group, Boca Raton, Florida, USA.

Pan, Z. and McHugh, T.H. (2006) Novel infrared dry-blanching (IDB), infrared blanching, and infrared drying technologies for food processing. US Patent 20,060,034,981.

Pan, Z., Shih, C., McHugh, T.H. and Hirschberg, E. (2008) Study of *banana* dehydration using sequential infrared radiation heating and freeze-drying. *Food Science and Technology*, **41**, 1944–1951.

Pathare, P.B. and Sharma, G.P. (2006) Effective moisture diffusivity of onion slices undergoing infrared convective drying. *Biosystems Engineering*, **93**, 285–291.

Person, N.K. and Sorenson, J.W. (1962) Drying hay with infrared radiation. *Agricultural Engineering*, **43**, 204–237.

Ponne, C.T., Baysal, T. and Yuksel, D. (1994) Blanching leafy vegetables with electromagnetic energy. *Journal of Food Science*, **59**, 1037–1041.

Ramaswamy, H.S. and Marcotte, M. (2005) *Food Processing: Principles and Applications*. CRC Press, Taylor & Francis Group, Boca Raton, Florida, USA.

Ranjan, R., Irudayaraj, J. and Jun, S. (2002) Simulation of three-dimensional infrared drying using a set of three-coupled equations by the control volume method. *Transactions of American Society of Agricultural Engineering*, **45**, 1661–1668.

Rosenthal, I., Rosen, B. and Berstein, S. **(**1996) Surface pasteurization of cottage cheese. *Milchwissenschaft*, **51**, 198–201.

Ruiz Celma, A., López-Rodríguez, F. and Cuadros Blázquez, F. (2009) Experimental modelling of infrared drying of industrial grape by-products. *Food and Bioproducts Processing*, **87**, 247–253.

Sakai, N. and Hanzawa, T. (1994) Applications and advances in far-infrared heating in Japan. *Trends in Food Science and Technology*, **5**, 357–362.

Sakai, N. and Mao, W. (2006) Infrared heating. In: *Thermal Food Processing*, Sun, De Wen (ed.). CRC Press, Boca Raton, Florida, USA.

Sakiyan, O., Sumnu, G., Sahin, S. and Meda, V. (2007) The effect of different formulations on physical properties of cakes baked with microwave and near infrared microwave combinations. *Journal of Microwave Power and Electromagnetic Energy*, **41**(1), 20–26.

Sakiyan, O., Sumnu, G., Sahin, S., Meda, V., Koksel, H. and Chang, P. (2011) A study on degree of starch gelatinization in cakes baked in three different ovens. *Food and Bioprocess Technology*, **4**, 1237–1244.

Sawai, J., Sagara, K., Hashimoto, A., Igarashi, H. and Shimizu, M. (2003) Inactivation characteristics shown by enzymes and bacteria treated with far-infrared radiative heating. *International Journal of Food Science and Technology*, **38**, 661–667.

Sandu, C. (1986) Infrared radiative drying in food engineering: a process analysis. *Biotechnology Progress*, **2**, 109–119.

Schroeder, H.W. and Rosberg, D.W. (1961) Infrared drying of rough rice. *Rice Journal*, **63**, 3–5.

Sharma, G.P., Verma, R.C. and Pathare, P.B. (2005a) Thin-layer infrared radiation drying of onion slices. *Journal of Food Engineering*, **67**, 361–366.

Sharma, G.P., Verma, R.C. and Pathare, P.B. (2005b) Mathematical modeling of infrared radiation thin layer drying of onion slices. *Journal of Food Engineering*, **71**, 282–286.

Shi, J., Pan, Z., McHugh, T.H., Wood, T., Zhu, Y., Avena-Bustillos, E.J. and Hirschberg, E. (2008) Effect of berry size and sodium hydroxide pretreatment on the drying characteristics of blue berries under infrared radiation heating. *Journal of Food Science*, **73**, E259–E265.

Shyu, Y.S., Sung, W.C., Chang, M.H. and Hwang, J.Y. (2008) Effect of far-infrared oven on the qualities of bakery products. *Journal of Culinary Science and Technology*, **6**, 105–118.

Skjöldebrand, C. and Andersson, C. (1989) A comparison of infrared bread baking and conventional baking. *Journal of Microwave Power and Electromagnetic Energy*, **24**, 91–101.

Staack, N., Ahrne, L., Borch, E. and Knorr, D. (2008a) Effect of infrared heating on quality and microbial decontamination in paprika powder. *Journal of Food Engineering*, **86**, 17–24.

Staack, N., Ahrne, L., Borch, E. and Knorr, D. (2008b) Effect of temperature, pH and controlled water activity on inactivation of spore of *Bacillus cereus* in paprika powder by near-IR radiation. *Journal of Food Engineering*, **89**, 319–324.

Sumnu, G., Sahin, S. and Sevimli. M. (2005) Microwave, infrared and infrared-microwave combination baking of cakes, *Journal of Food Engineering,* **71**, 150–155.

Sumnu, G., Datta, A.K., Sahin, S., Keskin, S.O. and Rakesh, V. (2007) Transport and related properties of breads baked using various heating modes. *Journal of Food Engineering*, **78**, 1382–1387.

Tanaka, F., Verboven, P., Scheerlinck, N., Morita, K., Iwasaki, K. and Nicolaï, B. (2007) Investigation of far-infrared radiation as an alternative technique for surface decontamination of strawberry. *Journal of Food Engineering*, **79**, 445–452.

Therdthai, N., Zhou, W. and Adamczak, T. (2002) Optimization of the temperature profile in bread baking. *Journal of Food Engineering*, **55**, 41–48.

Uysal, N., Sumnu, G. and Sahin, S. (2009) Optimization of microwave-infrared roasting of hazelnut. *Journal of Food Engineering*, **90**, 255–261.

van Zuilichem, D.J., Vant Reit, K. and Stolp, W. (1986) An overview of new infrared radiation processes for various agricultural products. *Food Engineering Process Applications*, **1**, 595–610.

Wade, P. 1987. Biscuit baking by near-infrared radiation, *Journal of Food Engineering*, **6**, 165–175.

Wang, J. and Sheng, K. (2006) Far-infrared and microwave drying of peach. *LWT-Food Science and Technology*, **39**, 247–255.

Yagi, S. and Kunii, D. (1951) Infrared drying characteristics of granular or powdery materials. *Kagaku Kikai*, **15**, 108–123.

Yang, J., Bingol, G., Pan, Z., Brandl, M.T., McHugh, T.H. and Wang, H. (2010) Infrared heating for dry-roasting and pasteurization of almonds. *Journal of Food Engineering*, **101**, 273–280.

Yang, J., Pan, Z., Takeoka, G., Mackey, B., Bingol, G., Brandl, M.T., Garcin, K., McHugh, T.H. and Wang, H. (2013) Shelf-life of infrared dry-roasted almonds. *Food Chemistry*, **138**(1), 671–678.

Zhu, Y. and Pan, Z. (2009) Processing and quality characteristics of apple slices under simultaneous infrared dry-blanching and dehydration with continuous heating. *Journal of Food Engineering*, **90**, 441–452.

Zhu, Y., Pan, Z., McHugh, T.H. and Barrett, D. (2010) Processing and quality characteristics of apple slices processed under simultaneous infrared dry-blanching and dehydration with intermittent heating. *Journal of Food Engineering*, **97**, 8–16.

20

Application of Radiowave Frequency in Food Processing

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20.1 Introduction

New developments in novel heating of foods often focus on assessment of the processing time, evaluation of heating uniformity, impact on quality attributes of the final products and prediction of energy efficiency. Studies of electromagnetic heating (electroheating) have a considerable portion among novel heating applications. Electroheating can be subdivided into direct electroheating, where an electrical current is applied directly (e.g. ohmic heating (OH)) or indirect electroheating (e.g. microwave (MW) or radio frequency (RF) heating) where electrical energy is converted into electromagnetic radiation, which subsequently generates heat within a product.

The potential use of this technology for food processing was recognized after 1950. It was first described as 'electric heat' and described how it is produced and suggested possible applications for food processing. This early effort employed RF energy for applications such as cooking meat products, heating bread and dehydration and blanching of vegetables. However, it was not commercialized due to high operating costs. By the 1960s, studies focused

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on defrosting of frozen products. These resulted in commercial production lines (Jason and Sanders, 1962a, 1962b). Demeczky (1974) also showed that RF application in juice processing (peach, quince and orange juices) had better bacteriological and sensory qualities compared to the conventional methods. In the late 1980s, the next generation of commercial RF applications began in post-bake drying of cookies and snack foods (Mermelstein, 1998; Rice, 1993). In the 1990s, RF pasteurization was studied with attempts to improve energy efficiency and technical problems such as run-away heating were solved (Zhao *et al.*, 2000). This, in turn, led to the RF requirement of RF applicator modifications and knowledge of dielectric properties at RF frequencies (Zhang, Lyng and Brunton 2006; Zhang *et al.*, 2004; Laycock, Piyasena and Mittal, 2003). There have been a number of review studies for RF use in food processing. Zhao *et al.* (2000) indicated major engineering challenges. Piyasena *et al.* (2003) reported the main industrial applications and reported the dielectric properties for a range of food products, while Marra *et al.* (2009) reviewed the achievements in the field of meat processing and post-harvest treatments of fruits.

20.2 Principles of RF processing

This section briefly overviews the RF heating mechanism. The physical theory of RF is found in Jones and Rowley (1997). The RF portion of the electromagnetic spectrum occupies between 1 and 300 MHz although the main frequencies for industrial heating lie in the range of 10 to 50 MHz (Tang, Wang and Chan, 2005). Within the latter range, only the selected frequencies (13.56 ± 0.00678, 27.12 ± 0.16272 and 40.68 ± 0.02034 MHz) are permitted for industrial, scientific and medical applications. RF energy is generated by a triode valve and applied via a pair of electrodes (Rowley, 2001). In the most common parallel plate RF system, one of these electrodes (the bottom electrode) is grounded to set up as a capacitor to store electrical energy. Jones and Rowley (1997) reviewed two other configuration types: fringe-field applicators (consisting of a series of bar, rod or narrow plate electrodes, suitable for heating or drying thin layers (< 10 mm)) and staggered through-field applicators (consisting of rod or tube shaped electrodes staggered on either side of a belt, suitable for heating products of intermediate thickness). When an alternating electrical field is applied, positive ions in the material move towards negative regions of the electric field and negative ions move towards positive regions of the field (Buffler, 1993). The movement of ions in this fashion is often referred to as ionic depolarization. Heating occurs because this field is not static, with polarity continually changing at high frequencies of the electromagnetic region (e.g. 27.12 MHz for RF or 2450 MHz for MW). In addition to movement of ions, dipolar molecules such as water attempt to align themselves appropriately with changing polarity (dipole rotation).

This movement also causes friction between molecules, leading to heat generation. While MW and RF heating are both referred to as dielectric heating methods, ionic depolarization tends to be the dominant heating mechanism at lower frequencies encountered in the RF range. On the other hand, both ionic depolarization and dipole rotation can follow dominant loss mechanisms at frequencies near the MW frequency range (i.e. 400–3000 MHz) depending upon the moisture and salt content within a product. The mathematical background required for possible design studies is available (Romano and Marra, 2008; Marra *et al.*, 2007; Jones and Rowley, 1997).

Conventional heating can be performed on a batch or continuous basis, and the heating mechanism applied can be conduction (e.g. in solid such as meat), convection (e.g. in liquid such as milk) or a mix of convection and conduction in products that display broken heating curves (e.g. some starch containing soups). The net effect is that overheating in the outer region occurs due to heating for a longer time to ensure the interior target temperature. In contrast, in electroheating (e.g. OH, MW and RF) heat is generated volumetrically within the material by either an alternating electrical current (as in OH) or electromagnetic radiation (formed by the conversion of electrical energy to electromagnetic radiation at MW or RF frequencies. During MW heating, waves (generated by a magnetron) pass via a waveguide into an oven cavity. In Ohmic heating, the product is placed in direct contact with a pair of electrodes through which generally a low-frequency (traditionally 50 or 60 Hz) alternating current is passed. Rowley (2001) reviewed the methods to produce and transmit RF power into materials and categorized the RF systems as 'conventional' or more recently introduced '50 Ω' systems.

20.3 Use of RF heating in food processing

20.3.1 Post-harvest treatment for agricultural commodities

Traditional fumigation agents such as methyl bromide are replaced by RF radiation for disinfection of fruits and nuts. Researchers have performed extensive studies on this topic (Hansen *et al.*, 2006a, 2006b; Monzon *et al.*, 2006; Wang *et al.*, 2006a, 2003a, 2002; Birla *et al.*, 2004; Mitcham *et al.*, 2004; Ikediala *et al.*, 2002; Tang *et al.*, 2000), including discussion on scaling-up and the design of RF systems (Wang *et al.*, 2007a, 2007b, 2006b). A study describing the intrinsic thermal mortality of insect pests demonstrated the potential to develop high-temperature–short-time thermal treatments to control codling moth in fruits (Tang *et al.*, 2000). When selected fruits and insect larvae were examined, loss factors of common pest insects at the RF range were found to be clearly larger than that of nuts (Wang *et al.*, 2003b). These results suggest a possible differential and a faster heating process of

insects during an RF process. Wang *et al.* (2002) processed walnut loads by means of a RF applicator (27 MHz–12 kW) to heat up to temperatures lethal to potential insect pests. Heating walnuts to 55 °C or higher resulted in 100% mortality of fifth instar navel orangeworm (*Amyelois transitella*), the most resistant among the possible three insect pests (*Cydia pomonella*, *Plodia interpunctella* and *Juglans regia*). Mitcham *et al.* (2004) conducted a follow-up study for a heavier walnut load in terms of heating uniformity. They concluded that moisture content was an important factor affecting heating rates. Birla *et al.* (2004) developed a lab-scale fruit-mover capable of continuously rotating the fresh fruit (with an orange and an apple as test samples) via a series of water jets to achieve uniform RF heating. They found that apples with typical geometry (oval and dimples on ends) showed more temperature variation compared to the almost spherical oranges. Some of the previous studies applied RF heating to fresh apples and sweet cherry, claiming the effectiveness of RF treatment for pest control with no major changes in the quality attributes. The impact on orange quality (weight loss, loss of firmness, colour change, total soluble solids, acidity and change in volatiles) has been assessed for RF treatments designed to control Mediterranean fruit flies. Monzon *et al.* (2007) evaluated the impact of RF heating to control Mexican fruit fly larvae on *Fuyu persimmons* fruit and found that the treatments used (48–52 °C followed by holding times of 0.5 to 18 min) had no significant effect on firmness, soluble solids content, titratable acidity or product weight loss. Wang *et al.* (2007b, 2006b) also proposed considerations about designing and scaling-up plants and processes for industrial-scale post-harvest RF treatment of walnuts with a continuous system characterized by high power (up to 25 kW) and high product load (up to 187 kg) moving on a belt. An effective RF treatment protocol has been developed for almonds for heating uniformity and occurrence of RF differential heating of insects. To prepare industrial-scale RF treatments for insect control in lentils, a treatment protocol was designed to provide 100% cowpea weevil mortality with a combined RF and forced hot-air treatment. Sisquella *et al.* (2013) studied the RF treatment at 27.12 MHz with fruits immersed in water to control brown rot in peaches and nectarines artificially inoculated with *M. fructicola*.

20.3.2 Meat processing

Laycock, Piyasena and Mittal (2003) compared the heating rate, time–temperature profiles and quality of three meat products (ground, comminuted and non-comminuted muscle) cooked in a water bath or by a 1.5 kW and 27.12 MHz RF heater (a cylindrical chamber set between two electrodes wrapped around the RF applicator). The results indicated that the cooking time was reduced by up to 1/25 and the surface of the RF cooked products became heated at a faster rate than the centre, with differences of 10–20 °C.

RF cooked samples had lower juice losses and were acceptable in terms of colour and water-holding capacity. Orsat *et al.* (2004) pasteurized vacuum packaged ham slices in an RF applicator (600 W at 27.12 MHz) and concluded that RF heating, coupled with appropriate packaging, could improve the storability of hams by decreasing their bacterial load, reducing moisture loss and maintaining an overall product sensory and quality acceptance.

The most extensive investigations on the processing and the quality of RF cooked meats were published between 2004 and 2007 (Lyng *et al.*, 2007; Zhang, Lyng and Brunton, 2006; McKenna *et al.*, 2006), where a 27.12 MHz RF applicator with a maximum power output of 600 W was used. These studies developed systems for RF cooking of cased meat samples to avoid arcing of the casings. They examined quality attributes of the RF cooked products including yield, texture, colour and flavour. In comminuted products, no yield differences were noted while for non-comminuted products (i.e. ham and beef), RF cooking had higher yields than steam cooked products (1–1.5% for ham and 4–6% for beef). Zhang, Lyng and Brunton (2004) developed an optimized cooking protocol for pasteurizing meat emulsion samples where reductions of up to 79% in pasteurization time were noted compared to an equivalent steam cooked sample. Recently, Byrne *et al.* (2010) studied radio frequency heating of comminuted meats considering the reduction of vegetative cells and spores of *Bacillus cereus* and *Clostridium perfringens*.

20.3.3 Drying and post-baking

Starting from the 1940s, RF drying has been used in drying of food materials. Ptasznik, Zygmunt and Kudra (1990) developed a simulation model for RF-assisted convective drying for the seed quality of broad bean. Murphy, Morrow and Besley (1992) studied drying of alfalfa using a combination of RF power at 27 MHz with heated forced air. Jumah (2005) prepared a theoretical analysis of simultaneous heat and mass transfer in the RF-assisted fluidized bed drying of corn particulates. The model was an effective tool for computer-aided optimization of RF-enhanced fluidized bed drying processes.

The main problem was in the bakery industry, where there was a lack of uniformity in the product moisture content. RF–post-baking processes can solve this problem. It was possible to add an RF drying unit immediately after a conventional baking oven to increase production and savings in floor space. RF drying provides a more uniform moisture content in final products and reduces the possibility of mould growth, which consequently increases the shelf-life (Mermelstein, 1998).

20.3.4 Other applications

Rapid heating and high penetration of RF energy in liquid foods makes it an alternative method of processing under continuous flow conditions. Heating

of starch and of guar gum dispersions was investigated (Awuah, Ramaswamy and Piyasena, 2002) with the objectives of evaluating the effect of the system (flow rate and RF power) and product parameters (starch or guar concentration, pH, added salt and sugar) on temperature change across the RF applicator (1.5 kW, 27.12 MHz) tube and to develop correlations for estimating the exit temperature for the considered samples. Awuah *et al.* (2005) used a 2 kW, 27.12 MHz RF applicator to find suitable conditions for inactivation of both *Listeria* and *Escherichia coli* in milk under continuous laminar flow conditions. Zhong, Sandeep and Swartzel (2003) reported a uniform heating pattern during the continuous flow of tap water in a 30 kW, 40.68 MHz RF applicator. On the other hand, 1% carboxymethylcellulose (CMC) dispersion in the same system exhibited higher temperature values at the tube inner walls. Wang *et al.* (2003a) compared RF heating (6 kW, 27.12 MHz) to conventional retort processes for sterilizing trays of macaroni and cheese to assess heating uniformity using a model food (whey protein gel charged with a chemical marker, M-1). They found the RF treated samples to be much closer in colour and flavour to the control sample, while retorted samples were darker in appearance and had a more burnt flavour. Another interesting application was proposed by Zhong, Sandeep and Swartzel (2004), where RF heating (30 kW, 40.68 MHz) was used to process carrot and potato cubes using a 1% CMC dispersion as a carrier. Based on thermal images captured by an infrared camera, they observed that a small gradient of temperature inside the carrot and potato cubes became heated for a short residence time. A 600 W, 27.12 MHz RF applicator was used by Orsat *et al.* (2001) to improve and extend the storability of vacuum-packaged carrots. Despite the fact that the quality of the RF treated samples was higher than in either control (processed with chlorinated water) or hot-water treated carrot samples, they suggested that RF treatment should be considered as a part of an integrated approach, including proper packaging and adequate refrigeration. Cserhalmi *et al.* (2001) had also reported successful reduction of pungency of yellow mustard seed after treatment in a 10 kW, 13.5 MHz RF system. Nelson *et al.* (2003) evaluated RF as a method to reduce *Salmonella*, *Escherichia coli* O157:H7 and *Listeria monocytogenes* contamination in alfalfa seeds. Short RF exposures (several seconds) produced reductions in the target organism without adverse effects on seed germination. However, extending RF exposure to produce a desired level of microbial reduction had an adverse effect on germination. In contrast to the aforementioned studies, Schuster-Gajzágó *et al.* (2006) exposed white mustard seed to RF with the intention of inactivating the endogenous enzyme myrosinase responsible for the development of a pungent sharp flavour. Ahmed *et al.* (2007) studied rheological and gelation characteristics of RF heated egg white dispersions. RF heated protein dispersions produced stronger gels in particular. Gao *et al.* (2011) used RF treatment to reduce *S. enteritidis* and evaluated the almond quality. Liu *et al.* (2013) studied heating uniformities of the

pre-packaged bread loaf using combined radio frequency (RF) and hot-air treatments to reduce mould growth.

20.4 Factors influencing RF heating processes

20.4.1 Dielectric properties

A review of dielectric property measurement techniques was provided by Regier and Schubert (2005). Dielectric properties of food materials can be divided into two parts, known as permeability and permittivity (ϵ). Permeability values for food materials are generally similar to that of free space and are not assumed to contribute to heating (Zhang and Datta, 2001). However, the permittivity reported in terms of dielectric constant (ϵ') and loss factor (ϵ'') influences the heating rates. The ϵ' and ϵ'', the real and imaginary parts, respectively, of ϵ are given by: $\epsilon = \epsilon' - j\epsilon''$. Here, ϵ' is a characteristic for any material and a measure of the polarizing effect from the applied electric field (i.e. how easily the medium is polarized). It also indicates the capacity to absorb, transmit and reflect energy from the electric portion of the electrical field. The term ϵ'' measures the amount of energy that is lost from the electrical field related to how the energy from a field is absorbed and converted to heat (Engelder and Buffler, 1991). In addition, electrical conductivity ($\sigma = 2\pi f\epsilon''$) indicates the ability of a material to conduct an electric current. In a dielectric food system, σ is related to ionic depolarization and contributes to ϵ''. The tangent of dielectric loss angle ($\tan\delta = \epsilon''/\epsilon'$), on the other hand, is called the loss tangent or dissipation (power) factor of the material. Another important property, penetration depth (d_p) is defined as the depth in a material where the energy of a plane wave propagating perpendicular to the surface has decreased to 1/e (1/2.71828) of its surface value (Bengtsson and Risman, 1971):

$$d_p = \frac{C}{2\pi f\sqrt{2\epsilon'}\sqrt{\sqrt{1+(\tan\delta)^2}-1}} \tag{20.1}$$

where C is the speed of propagation of waves in a vacuum (3×10^8 m/s). When $\tan\delta$ is low ($<< 1$), the penetration depth is well described by

$$d_p = \frac{C}{2\pi f\sqrt{\epsilon'}\tan\delta} = \frac{4.47\times10^7}{f\sqrt{\epsilon'}\tan\delta} \tag{20.2}$$

20.4.2 Factors influencing dielectric properties of foods

Dielectric properties play an important role in electroheating processes. These properties are influenced by the moisture content, frequency of applied alternating field, temperature and chemical composition of the material. In relation

to composition, Nelson and Datta (2001) stated that the dielectric properties are dependent on the chemical composition and especially the presence of mobile ions and permanent dipole moments associated with water. Published dielectric data are available on fruits, mashed potato, whey protein gel, macaroni, cheese, egg and salmon. Results show that ϵ'' of whey protein products, macaroni and cheese increased sharply with increasing temperature at 27 and 40 MHz. The ϵ' of mashed potato increased with temperature at 27 MHz but stayed stable at 40 MHz while the ϵ'' increased with temperature at both 27 and 40 MHz. Moisture content did not affect the dielectric properties while added salt had a significant influence on both dielectric properties, which in turn influenced d_p. Other researchers conducted investigations at 27.12 MHz on the dielectric properties of meat, meat products and ingredients used in the manufacture of meat products. The results showed that most of the additives that have free ions can change the dielectric properties of the final product and in turn influence the RF heating process. Farag *et al.* (2008) evaluated the dielectric properties of three different beef meat blends (lean, fat and 50:50 mixture) over a temperature range from −18 to +10 °C. In the region of thawing (−3 to −1 °C), ϵ' and ϵ'' values at 27.12 MHz were significantly higher ($P < 0.05$) than at other measured temperatures for the three blends. The composition also significantly influenced ($P < 0.05$) the measured dielectric properties at all temperatures used, and thus the heating pattern during thawing.

20.5 Computer simulation of RF heating in food processing

Starting from mid 1990s, a number of studies discussed mathematical modelling and computer simulation of RF systems (Birla, Wang and Tang, 2008; Romano and Marra, 2008; Marra *et al.*, 2007; Chan, Tang and Younce, 2004; Yang, Zhao and Wells, 2003; Neophytou and Metaxas, 1998). Neophytou and Metaxas (1998) presented a three-dimensional FEM model for characterization of electric fields within RF applicators. To establish the validity of electrostatic conditions, they solved both Laplace (for electrostatic conditions) and wave equations. The authors concluded that electrostatic conditions (and the Laplace equation) can be assumed only in the case of a very small experimental size applicator. Yang, Zhao and Wells (2003) proposed the simulation of heating performance of radish and alfalfa sprout seeds packed inside rectangular seed boxes during RF heating to evaluate the electric field distribution. They reported discrepancies between simulated and experimental results, especially at the edges of the box. Further, the understanding of RF applicators by computer simulations was proposed by Chan, Tang and Younce (2004), where the method proposed by Neophytou and Metaxas (1999) was followed by adding a means of excitation to the tank oscillatory circuit with an external, properly positioned, coaxial source.

For applying an electro-quasi-static hypothesis for a small cavity, Marra *et al.* (2007) solved coupled electromagnetic and heat transfer equations, using a commercial FEM-based software, for a cylindrical shaped meat batter placed between the electrodes of a 600 W RF system. This work demonstrated that the electro-quasi-static hypothesis can be applied successfully to lab-scale RF apparatus, and it may be used for further investigation on RF heating by means of computer simulation (Birla, Wang and Tang, 2008; Romano and Marra, 2008).

20.6 Conclusions

In recent years, the interest in RF heating technology has increased significantly and its applications have increased by several folds in the food and material processing industries. The present chapter discusses the various aspects of RF heating technology in concert with its applications on the quality of processed foods. The rapid heating method of RF offers considerable advantages over conventional slow heating processes, rendering significant applications in different food products. The physical factors such as shape, geometry and product position, as well as dielectric properties, are the fundamental factors that affect the RF heating systems. Basic understanding of these factors and experimental studies of different materials processed by this novel technology are the crucial points to deal with in order to increase the versatility and applicability of RF technology. This chapter is believed to address what has been done so far and what has to be done in the future, and to extend the use of RF technology in a variety of food materials.

References

Ahmed, J., Ramaswamy, H.S., Alli, I. and Raghavan, V.G.S (2007) Protein denaturation, rheology and gelation characteristics of radiofrequency heated egg white dispersions. *International Journal of Food Properties*, **10**, 145–161.

Awuah, G.B., Ramaswamy, H.S. and Piyasena, P. (2002) Radio frequency (RF) heating of starch solutions under continuous flow conditions: effect of system and product parameters on temperature change across the applicator tube. *Journal of Food Process Engineering*, **25**(3), 201–223.

Awuah, G.B., Ramaswamy, H.S., Economides, A. and Mallikarjuanan, K. (2005) Inactivation of *Escherichia coli* K-12 and *Listeria innocua* in milk using radio frequency (RF) heating. *Innovative Food Science and Emerging Technologies*, **6**(4), 396–402.

Bengtsson, N.E. and Risman, P.O. (1971) Dielectric properties of foods at 3 GHz as determined by cavity perturbation technique. *Journal of Microwave Power*, **6**(2), 107–123.

Birla, S.L., Wang, S. and Tang, J. (2008) Computer simulation of radio frequency heating of model fruit immersed in water. *Journal of Food Engineering*, **84**, 270–280.

Birla, S.L., Wang, S., Tang, J. and Hallman, G. (2004) Improving heating uniformity of fresh fruit in radio frequency treatments for pest control. *Postharvest Biology and Technology*, **33**(2), 205–217.

Buffler, C.R. (1993) Dielectric properties of foods and microwave materials. In: *Microwave Cooking and Processing*. Van Nostrand Reinhold, New York, USA, pp. 46–69.

Byrne, B., Lyng, J.G., Dunne, G. and Bolton, D.J. (2010) Radio frequency heating of comminuted meats – considerations in relation to microbial challenge studies. *Food Control*, **21**, 125–131.

Chan, T.V.C.T., Tang, J. and Younce, F. (2004) 3-Dimensional numerical modeling of an industrial radio frequency heating system using finite elements. *Journal of Microwave Power and Electromagnetic Energy*, **39**(2), 87–106.

Cserhalmi, Zs., Márkus, Zs., Czukor, B., Baráth, Á. and Tóth, M. (2001) Physico-chemical properties and food utilization possibilities of RF-treated mustard seed. *Innovative Food Science and Emerging Technologies*, **1**(4), 251–254.

Demeczky, M. (1974) Continuous pasteurisation of bottled fruit juices by high frequency energy. *Proceedings of IV International Congress on Food Science and Technology*, **IV**, 11–20.

Engelder, D.S. and Buffler, C.R. (1991) Measuring dielectric properties of food products at microwave frequencies. *Microwave World*, **12**(2), 2–11.

Farag, K., Lyng, J.G, Morgan, D.J. and Cronin, D.A. (2008) Dielectric and thermophysical properties of different beef meat blends over a temperature range of −18 to +10 °C. *Meat Science*, **79**(4), 740–747.

Gao, M., Tang, J., Villa-Rojas, R., Wang, Y. and Wang, S. (2011) Pasteurization process development for controlling *Salmonella* in in-shell almonds using radio frequency energy. *Journal of Food Engineering*, **104**, 299–306.

Hansen, J.D., Drake, S.R., Heidit, M.L., Watkins, M.A., Tang, J., and Wang, S. (2006a) Radio frequency–hot water dips for postharvest codling moth control in apples. *Journal of Food Processing and Preservation*, **30**, 631–642.

Hansen, J.D., Drake, S.R., Watkins, M.A., Heidit, M.L., Anderson, P.A. and Tang, J. (2006b) Radio frequency pulse application for heating uniformity in postharvest codling moth (*Lepidoptera tortricidae*) control of fresh apples (*Malus domestica* borkh.). *Journal of Food Quality*, **29**, 492–504.

Ikediala, J.N., Hansen, J.D., Tang, J., Drake, S.R. and Wang, S. (2002) Development of a saline water immersion technique with RF energy as a postharvest treatment against codling moth in cherries. *Postharvest Biology and Technology*, **24**(1), 25–37.

Jason, A.C. and Sanders, H.R. (1962a) Dielectric thawing of fish. I. Experiments with frozen herrings. *Food Technology*, **16**(6), 101–106.

Jason, A.C. and Sanders, H.R. (1962b) Dielectric thawing of fish. II. Experiments with Frozen White Fish. *Food Technology*, **16**(6), 107–112.

Jones, P.L. and Rowley, A.T. (1997) In: *Industrial Drying of Foods* (Baker C.G.J., ed), Chapter 8. Blackie Academic and Professional, London, England.

Jumah, R. (2005) Modelling and simulation of continuous and intermittent radio frequency-assisted fluidized bed drying of grains. *Food and Bioproducts Processing*, **83**, 203–210.

Laycock, L., Piyasena, P. and Mittal, G.S. (2003) Radio frequency cooking of ground, comminuted and muscle meat products. *Meat Science*, **65**(3), 959–965.

Liu, Y., Wang, S., Mao, Z., Tang, J. and Tiwari, G. (2013) Heating patterns of white bread loaf in combined radio frequency and hot air treatment. *Journal of Food Engineering*, **116**, 472–477.

Lyng, J.G., Cronin, D.A., Brunton, N.P., Li, W. and Gu, X. (2007) An examination of factors affecting radio frequency heating of an encased meat emulsion. *Meat Science*, **75**(3), 470–479.

Marra, F., Lyng, J., Romano, V. and McKenna, B. (2007) Radio-frequency heating of foodstuff: solution and validation of a mathematical model. *Journal of Food Engineering*, **79**(3), 998–1006.

McKenna, B.M., Lyng, J., Brunton, N. and Shirsat, N. (2006) Advances in radio frequency and Ohmic heating of meats. *Journal of Food Engineering*, **77** (2), 215–229.

Mermelstein, N.H. (1998) Microwave and radio frequency drying. *Food Technology*, **52**(11), 84–86.

Mitcham, E.J., Veltman, R.H., Feng, X., de Castro, E., Johnson, J.A., Simpson, T.L., Biasi, W.V., Wang, S. and Tang, J. (2004) Application of radio frequency treatments to control insects in in-shell walnuts. *Postharvest Biology and Technology*, **33**(1), 93–100.

Monzon, M.E., Biasi, B., Simpson, T.L., Johnson, J., Feng, X., Slaughter, D.C. and Mitcham, E.J. (2006) Effect of radio frequency heating as a potential quarantine treatment on the quality of 'Bing' sweet cherry fruit and mortality of codling moth larvae. *Postharvest Biology and Technology*, **40**(2), 197–203.

Monzon, M. E., Biasi, B., Mitcham, E. J., Wang, S., Tang, J. and Hallman, G.J. (2007) Effect of radiofrequency heating on the quality of Fuyu persimmon fruit as a treatment for control of the Mexican fruit fly. *Horticulture Science*, **42**(1), 125–129.

Murphy, A, Morrow, R. and Besley, L. (1992) Combined radiofrequency and forced-air drying of alfalfa. *Journal of Microwave Power and Electromagnetic Energy*, **27**(4), 223–232.

Nelson, S.O. and Datta, A.K. (2001) Dielectric properties of food materials and electric field interactions. In: *Handbook of Microwave Technology for Food Applications* (Datta, A.K. and Anantheswaran, R.C., eds). Marcel Dekker, New York, USA.

Nelson, S.O., Lu, C.Y., Beuchat, L.R., and Harrison, M.A. (2003) Radio-frequency heating of alfalfa seed for reducing human pathogens. *Transactions of the ASAE*, **45**(6), 1937–1942.

Neophytou, R.I. and Metaxas, A.C. (1998) Combined 3D FE and circuit modeling of radio frequency heating systems. *Journal of Microwave Power and Electromagnetic Energy*, **33**(4), 243–262.

Neophytou, R.I. and Metaxas, A.C. (1999) Combined tank and applicator design of radio frequency heating systems. *IEE Proceedings – Microwave Antennas Propagation*, **146**(5), 311–318.

Orsat, V., Gariépy, Y., Raghavan, G.S.V. and Lyew, D. (2001) Radio-frequency treatment for ready-to-eat fresh carrots. *Food Research International*, **34**, 527–536.

Orsat, V., Bai, L., Raghavan, G.S.V. and Smith, J.P. (2004) Radio-frequency heating of ham to enhance shelf-life in vacuum packaging. *Journal of Food Process Engineering*, **27**, 267–283.

Piyasena, P., Dussault, C., Koutchma, T., Ramaswamy, H.S. and Awuah, G.B. (2003) Radio frequency heating of foods: principles, applications and related properties. A review. *Critical Reviews in Food Science and Nutrition*, **43**(6), 587–606.

Ptasznik W, Zygmunt, S. and Kudra T. (1990) Simulation of RF-assisted convective drying for seed quality broad bean. *Drying Technology*, **8**(5), 977–992.

Regier, M. and Schubert, H. (2005) Measuring delectric properties of foods. In: *The Microwave Processing of Foods* (Schubert, H. and Regier, M., eds). Woodhead Publishing, Cambridge, England, pp. 41–60.

Rice, J. (1993) RF technology sharpens bakery's competitive edge. *Food Processing*, **6**, 18–24.

Romano, V. and Marra, F. (2008) A numerical analysis of radio frequency heating of regular shaped foodstuff. *Journal of Food Engineering*, **84**, 449–457.

Rowley, A.T. (2001) Radio frequency heating. In: *Thermal Technologies in Food Processing* (Richardson, P., ed.). Woodhead Publishing, Cambridge, England, pp. 162–177.

Schuster-Gajzágó, I., Kiszter, A.K., Tóth-Márkus, M., Baráth, A., Márkus-Bednarik, Z. and Czukor, B. (2006) The effect of radio frequency heat treatment on nutritional and colloid-chemical properties of different white mustard varieties. *Innovative Food Science and Emerging Technologies*, **7**, 74–79.

Sisquella, M., Casals, C., Picouet, P., Viñas, I., Torres, R. and Usall, J. (2013) Immersion of fruit in water to improve radio frequency treatment to control brown rot in stone fruit. *Postharvest Biology and Technology*, **80**, 31–36.

Tang, J., Ikediala, J.N., Wang, S., Hansen, J.D. and Cavalieri, R.P. (2000) High-temperature–short-time thermal quarantine methods. *Postharvest Biology and Technology*, **21**, 129–145.

Tang, J., Wang, Y. and Chan, T.V.C.T. (2005) Radio frequency heating in food processing. In: *Novel Food Processing Technologies* (Gustavo Barbosa-Cánovas, G.V., Tapia, M.S. and Cano, M.P., eds). Marcel Dekker, New York, USA, pp. 501–524.

Wang, S., Tang, J., Johnson, J.A., Mitcham, E., Hansen, J.D., Cavalieri, R.P., Bower, J. and Biasi, B. (2002) Process protocols based on radio frequency energy to control field and storage pests in in-shell walnuts. *Postharvest Biology and Technology*, **26**(3), 265–273.

Wang, Y., Wig, T.D., Tang, J. and Hallberg, L.M. (2003a) Sterilization of foodstuffs using radio frequency heating. *Journal of Food Science*, **68**(2), 539–544.

Wang, S., Tang, J., Johnson, J.A., Mitcham, E., Hansen, J.D., Hallman, G., Drake, S.R. and Wang, Y. (2003b) Dielectric properties of fruits and insect pests as related to radio frequency and microwave treatments. *Biosystems Engineering*, **85**(2), 201–212.

Wang, S., Birla, S.L., Tang, J. and Hansen, J.D. (2006a) Postharvest treatment to control codling moth in fresh apples using water assisted radio frequency heating. *Postharvest Biology and Technology*, **40**(1), 89–96.

Wang, S., Tang, J., Sun, T., Mitcham, E.J., Koral, T. and Birla, S.L. (2006b) Considerations in design of commercial radio frequency treatments for postharvest pest control in in-shell walnuts. *Journal of Food Engineering*, **77**(2), 304–312.

Wang, S., Monzon, M., Johnson, J.A., Mitcham, E.J. and Tang, J. (2007a) Industrial-scale radio frequency treatments for insect control in walnuts. I: Heating uniformity and energy efficiency. *Postharvest Biology and Technology*, **45**, 240–246.

Wang, S., Monzon, M., Johnson, J.A., Mitcham, E.J. and Tang, J. (2007b) Industrial-scale radio frequency treatments for insect control in walnuts II: Insect mortality and product quality. *Postharvest Biology and Technology*, **45**, 247–253.

Yang, J., Zhao, Y. and Wells, J.H. (2003) Computer simulation of capacitive radio frequency (RF) dielectric heating on vegetable sprout seeds. *Journal of Food Process Engineering*, **26**, 239–263.

Zhang, H. and Datta, A.K. (2001) Electromagnetics of microwave heating: magnitude and uniformity of energy absorption in an oven. In: *Handbook of Microwave Technology for*

Food Applications (Datta, A.K. and Anantheswaran, R.C., eds). Marcel Dekker, New York, USA, pp. 1–28.

Zhang, L., Lyng, J.G. and Brunton, N.P. (2004) Effect of radio frequency cooking on texture, colour and sensory properties of a large diameter comminuted meat product. *Meat Science*, **68**(2), 257–268.

Zhang, L., Lyng, J.G. and Brunton, N.P. (2006) Quality of radio frequency heated pork leg and shoulder ham. *Journal of Food Engineering*, **75**(2), 275–287.

Zhang, L., Lyng, J.G., Brunton, N., Morgan, D. and McKenna, B. (2004) Dielectric and thermophysical properties of meat batters over a temperature range of 5–85 °C. *Meat Science*, **68**, 173–184.

Zhao, Y., Flugstad, B., Kolbe, E., Park, J.E. and Wells, J.H. (2000) Using capacitive (radio frequency) dielectric heating in food processing and preservation – a review. *Journal of Food Process Engineering*, **23**, 25–55.

Zhong, Q., Sandeep, K.P. and Swartzel, K.R. (2003) Continuous flow radio frequency heating of water and carboxymethylcellulose solutions. *Journal of Food Science*, **68**(1), 217–223.

Zhong, Q., Sandeep, K.P. and Swartzel, K.R. (2004) Continuous flow radio frequency heating of particulate foods. *Innovative Food Science and Emerging Technologies*, **5**(4), 475–483.

21

Application of Ultrasonics in Food Preservation and Processing

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21.1 Introduction

Technology and innovation are important aspects of food manufacturing along with the safety and quality characteristics of the final product and the environmental issues. The most important trends in the development of novel food processing technologies are relative to plant and process. The design of energy saving equipment is an important issue in food processing, since electricity represents the main input of the process; the reduction in the consumption of electricity leads to better environmental and cost performances. The food industries still use heat through thermal processing operations (pasteurization, sterilization, etc.) in order to guarantee the microbiological safety of products. These traditional heating methods rely on the generation of heat outside the product to be heated, by combustion of fuels or by an electric resistive heater, and its transference into the product mostly through conduction and convection mechanisms. These ways of processing are still limited due to considerable losses of heat on the surfaces of the equipment, reduction of heat transfer efficiency and thermal damage by overheating due to the time required to conduct sufficient heat into the thermal centre of foods. Another important aspect that must be taken into account is the quality attributes (flavour and odour, visual appearance, colour

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and texture, nutrition value and absence of additives). Many food ingredients and products are well known to be thermally sensitive and can be damaged/inactivated to chemical, physical and microbiological changes. Losses of some compounds, low production efficiency and time- and energy-consuming procedures such as prolonged heating and stirring may be encountered using these conventional food processing methods. These shortcomings have led to continuous industrial interest in developing alternatives – 'green and innovative' techniques in processing and preservation of food, which may be used to replace the severe heat-based methods that are commonly used.

Recent advances in the search for such nonthermal processing methods led researchers to investigate the application of ultrasound. Ultrasound is an example of new technology and its application in food processing may lead to both these areas undergoing an improvement. The applications for high power ultrasound (HPU) in food processing are numerous and include degassing, extractions, induction of oxidation/reduction reactions, nucleation for crystallization processes, cleaning of organic/inorganic surfaces and porous interior structures, reducing the particle size and variability in liquid dispersions and the defouling of filters. Also, ultrasound can pasteurize and preserve foods by inactivating many enzymes and microorganisms at mild temperature conditions, which can improve food quality in addition to guaranteeing stability and safety of foods.

21.2 Ultrasound mechanism

Ultrasonication is the application of high intensity sound waves at frequencies between 16 kHz and 100 MHz (Mason and Cordmas, 1996). The lowest frequency classification in the acoustic spectrum is infrasound, which has a frequency range less than about 20 Hz. Audible sound is what human beings hear and has an approximate frequency range between 16 Hz and 18 kHz. The ultrasound frequency range starts at a frequency of about 20 kHz (Figure 21.1). The ultrasound range can be divided into three different frequency ranges (Ashokkumar and Kentish, 2011):

- diagnostic ultrasound (1–10 MHz),
- high-frequency ultrasound having 100 kHz–1 MHz with low sound intensity (0.1 – 1 W/cm^2),
- low-frequency power ultrasound in the kHz range (20–100 kHz) with high sound intensity (10 – 1000 W/cm^2) .

Diagnostic ultrasound involves low-amplitude (higher frequency) propagation, which is concerned with the effect of the medium on the wave; it is commonly referred to as 'low-power' or 'high-frequency ultrasound'.

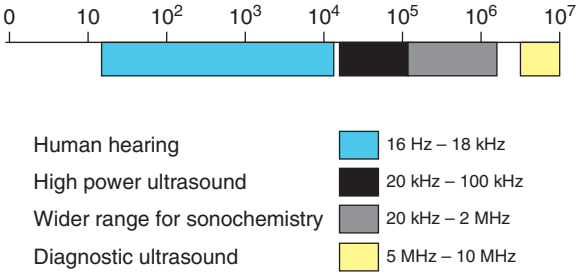

Figure 21.1 Range of ultrasound frequency (source: Leonelli and Mason, 2010. Reproduced with permission of Elsevier) *See plate section for colour version*

Typically, low-amplitude waves are used to measure the velocity and absorption coefficient of the wave in a medium in the 1–10 MHz range. Useful industrial applications include texture, viscosity and concentration measurements of many solid or liquid foods such as the composition determination of eggs, meats, fruits and vegetables, dairy and other products. High-frequency ultrasound involves ultrasound frequency in the range of 100 kHz–1 MHz with low sound intensity ($0.1 - 1\ W/cm^2$). Power ultrasound involves high-energy (low-frequency) waves known as 'power ultrasound'. Frequency in this case is between 20 and 100 kHz, which is used for food processing as pre-treatment, in extractions, freezing, drying, defoaming, cleaning, depolymerization, disaggregation, inactivation of microorganisms, etc. The beneficial use of sound is realized through its chemical, mechanical or physical effects on the process or product.

The characteristics of ultrasound waves (Patist and Bates, 2008) are as follows:

- Propagation. The propagation properties are generally used to describe quantitatively the propagation of ultrasound in materials, such as speed, impedance and attenuation.
- Frequency is inversely proportional to the bubble size; thus power ultrasound generates larger bubbles in the cavitation zone, resulting in higher temperatures and pressures.
- Amplitude must be delivered, maintained and monitored. Amplitude must be controlled under variable load conditions within the specifications.
- Absorption is a mechanism that represents that portion of the wave energy that is converted into heat, and scattering can be thought of as the portion that changes direction.
- Power (P), which is measured in watts (W), is the energy required to move the mechanical masses used to create cavitation in a liquid at a

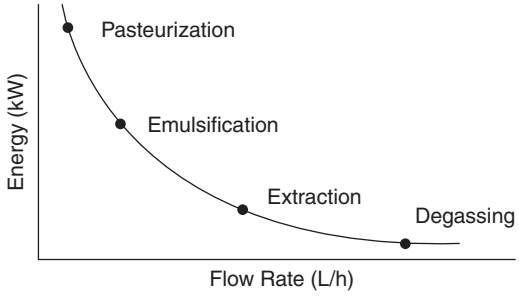

Figure 21.2 Graph of flow rate (L/h) and energy (kW) for ultrasound applications (source: Patist and Bates 2008. Reproduced with permission of Elsevier.)

specified amplitude of vibration, against a specified load at the fixed resonant frequency of the device:

$$P = m \times C_p \times \left(\frac{\partial T}{\partial t}\right)_{t=0} \tag{21.1}$$

Here, m is the mass of the sample (kg), C_p is the specific heat capacity of the medium (kJ/kg K), t is the time (s) and T is the temperature (K).

- Ultrasonic energy is expressed as energy input per volume of treated material (W/L). The energy input is the power output (W) and the flow rate (L/h) of the liquid through the ultrasonic processor, which relates to the time of exposure. Figure 21.2 shows the general relationship between ultrasound energy and flow rate for several ultrasonic applications.
- Intensity is a measure of the energy available per unit volume of liquid and is directly related to amplitude. Intensity refers to the power output per surface area of the sonotrode (W/cm^2):

$$AI = P/A \tag{21.2}$$

where AI is the ultrasonic intensity (W/cm^2) and A is the surface area of the probe (cm^2).

21.2.1 Cavitation

Power ultrasound (20–100 kHz) can provide the mechanical effect of cavitation in liquid systems, which can alter physical and chemical properties of food depending on the type of material involved. When ultrasound waves pass through a medium, a series of compression and rarefaction waves on the molecules of the medium are produced. This enforces a sinusoidal acoustic

pressure (Pa) in addition to the hydrostatic pressure acting on the medium (Soria and Villamiel, 2010). If a large negative pressure (sufficiently below ambient) is applied to the liquid so that the distance between the molecules exceeds the critical molecular distance necessary to hold the liquid intact, the liquid will break down and the cavitation bubbles will be formed (O'Brien, 2007). These bubbles are formed from the gas nuclei within the fluid and are distributed throughout the liquid. After a period of a few cycles, the bubbles will grow into a critical size, which makes them unstable and collapse violently. The collapsing bubbles will create energy accumulated hot spots, which can generate a high temperature (5000 K) and pressure (1000 atm), resulting in high shear energy zones and turbulence in the cavitation zone of the liquid (Chemat, Huma and Kamran Khan, 2011). The extent of cavitation is affected by energy, intensity, medium viscosity, surface tension, vapour pressure, nature and concentration of the dissolved gas, temperature and pressure of the treatment and the presence of solid particles. The formation of cavitation bubbles within a liquid is more difficult with increasing frequency from kHz to MHz. High frequencies form short cycles of compression and rarefaction ultrasonic waves, which cannot separate liquid molecules to form voids, and hence produce no cavitation. Therefore, most of the industrial ultrasound applications utilize the frequency range between 16 and 100 kHz in order to obtain the cavitation effect (Santos, Lodeiro and Capelo, 2009).

Two competing theories exist to explain the chemical effects due to cavitation – the *hot-spot theory* and the *electrical theory*. The hot-spot theory postulates that when the bubbles cavitate, localized hot spots are formed, which reach temperatures and pressures in excess of 5000 K and 1000 atm, respectively. Under these extreme conditions, chemical compounds can be degraded through three distinct pathways, such as (1) oxidation by hydroxyl radicals, (2) pyrolytic decomposition and (3) supercritical water oxidation (Hoffmann, Hua and Höchemer, 1996). The electrical theory postulates that an electrical charge is created on the surface of a cavitation bubble, forming enormous electrical field gradients across the bubbles, which are capable of bond breakage upon collapse.

Radical formation Ultrasound treatment leads to cavitation, microstreaming, rise in temperature and pyrolysis of water. During the implosion of cavitation bubbles, high local temperatures and pressures occur, leading to reaction in the surroundings. Free radicals are formed to initiate several chain reactions:

$$H_2O \rightarrow OH\cdot + H\cdot$$

$$OH\cdot + H\cdot \rightarrow H_2O$$

$$2OH\cdot \rightarrow H_2O_2$$

$$OH\cdot + OH\cdot \rightarrow H_2 + O_2$$

$$2H\cdot \rightarrow H_2$$
$$2OH\cdot \leftrightarrow O\cdot + H_2O$$
$$2O\cdot \leftrightarrow O_2$$
$$H_2O\cdot + 2H\cdot \rightarrow H_2O + H_2$$
$$H_2O\cdot + H_2O\cdot + O_2 \rightarrow 2H_2O_2$$
$$HO\cdot + H_2O \rightarrow H_2O_2 + H\cdot$$

Additional reactions in the presence of oxygen:

$$O_2 \rightarrow 2O\cdot$$
$$O_2 + O\cdot \rightarrow O_3$$
$$O_2 + H\cdot \rightarrow OOH\cdot \text{ (or } OH\cdot + O)$$
$$O\cdot + OOH\cdot \leftrightarrow OH\cdot + O_2$$
$$O\cdot + H_2O \rightarrow 2OH\cdot$$

Type of cavitation When driven by a sound field, the bubbles pulsate, which is termed stable cavitation (noninertial cavitation). Under stable cavitation, the bubbles pulsate over many cycles and remain physically intact, and the physical effects generated by these pulsations are characteristic of 'low energy' sources. Free radicals are produced by 'high energy' sonication effects and are associated with a phenomenon known as transient cavitation (results in the break-up of the bubble). This kind of cavitation resulting in 'high-energy' effects is termed inertial cavitation. Both the high gas temperatures and the gas shocks may generate free radicals (e.g. in aqueous solutions, H atoms, OH radicals and products such as hydrogen peroxide).

There are four types of cavitation based on the mode of generation: acoustic, optic, particle and hydrodynamic. In food processing industries, only acoustic and hydrodynamic cavitation is found to be efficient (Gogate and Kabadi, 2009) since they produce chemical or physical changes in the treated material.

21.2.2 Acoustic streaming

One of the acoustic streaming phenomena relevant to sonoporation is microstreaming, which is small-scale and boundary-associated acoustic streaming generated by a trapped oscillating bubble. Microstreaming is a significant acoustic phenomenon associated with cavitation, which occurs when the oscillating bubbles produce a vigorous circulatory motion, and thus set up strong eddy currents in the fluid surrounding them (Starritt, Duck and Humphrey, 1989).

21.2.3 Equipment producing ultrasound

The piezoelectric transducer The most common way of producing and detecting ultrasound utilizes the piezoelectric properties of certain crystalline materials, such as quartz. Thus, applying rapidly reversing charges to piezoelectric material causes fluctuations in its dimensions. This effect offers transmitting ultrasonic vibrations from the crystal section through whatever medium it might be in. There are four types of laboratory ultrasonic apparatus commercially available in general; these are whistle reactors, ultrasonic cleaning baths, probes and cup-horn devices. Cleaning baths and probes are usually operated at a fixed frequency dependent on the particular type of transducer, which is usually 20 kHz for common probe systems and 40 kHz for baths.

Ultrasound bath A cleaning bath consists of a stainless steel tank with transducers clamped to its base. One of the basic parameters in ultrasonic engineering is power density, which is defined as the electrical power delivered to the transducer divided by the transducer radiating surface area. An ultrasonic bath, which is a low-intensity device, uses a power intensity at the transducer face of about $1 - 5\ W/cm^2$ in modern piezoelectric transducers and operates at a frequency of 40 kHz.

Ultrasonic probes A typical ultrasonic processor consists of a generator, which transforms main voltage into high-frequency electrical energy at 20 kHz. This is fed to an ultrasonic converter element (called a piezoelectric transducer), which transforms supplied electrical energy to 20 kHz mechanical vibratory energy (Mason, 1998). A sonotrode (probe) delivers its energy on a specific zone, where cavitation is dramatically boosted. An ultrasound probe comprises different components such as power supply, a transducer (converter) to convert electrical power into mechanical vibrations, piezoelectric ceramic disks, electrodes and a sonotrode (the sole part of the system that can be exchanged). Sonotrodes come in all shapes and sizes depending on the intended use (it should be resonant at the operating frequency). The material from which it is constructed (mild steel, stainless steel, titanium alloy, ceramics) should be a compromise between the needs of ultrasound and those of the application. The tip is the radiating surface of the horn, which irradiates acoustic energy outwards. Tips may be either removable or integrated with the final output element. They differ in diameters. Optional parts are a booster (titanium or high-strength aluminum alloy), fitted between the transducer, and the ultrasonic tool and extender.

A continuous system (flow cells) offers in-line or continuous processing of large sample volumes. Flow cells are ideal for mixing and dispersing applications and can accommodate high flow rates. As the sample passes through the cavitation field, it is processed. The processed liquid exits the unit through an outlet port. The sample can be recirculated multiple times if necessary.

The degree of processing may be controlled by adjusting both the amplitude setting and the flow rate.

21.3 Application of ultrasound in food processing

The different applications of ultrasound in the food industries including their advantages are shown in Table 21.1.

21.3.1 Ultrasound and microbial inactivation

Ultrasonic pasteurization between temperatures of 40 and 50 °C has the potential of preserving the quality of many food products in terms of physicochemical properties, colour and flavour compared to conventional pasteurization techniques, which operate at much higher temperatures. Most microorganisms show greater sensitivity to ultrasound at increased temperatures over 50 °C (Villamiel and de Jong, 2000). However, some authors claim that it is possible to inactivate microorganisms even at a temperature of 40 °C (Herceg *et al.*, 2013, 2012; Herceg, Juraga, Sobota and Režek Jambrak, 2012). An elevated temperature weakens the bacterial membrane, which enhances the effect of cavitation due to the ultrasound. In particular, the use of high-power ultrasound has shown several advantages compared to heat pasteurization, such as minimization of flavour loss in juices, greater homogeneity and significant energy savings (Herceg *et al.*, 2012; Juraga *et al.*, 2011). In combination with heat, ultrasound can accelerate the rate of heat treatment of foods, thereby lessening the duration and intensity of thermal treatment and the resultant damage (Piyasena, Mohareb and McKellar, 2003). Many researchers have attempted to understand the mechanism played by ultrasound on the inactivation of microorganisms (Herceg *et al.*, 2013, 2012; Herceg, Juraga, Sobota and Režek Jambrak 2012; Bermudez-Aguirre, Mobbs and Barbosa-Canovas, 2011; Raso *et al.*, 1998). It can be explained by the phenomenon of acoustic cavitation and its physical, mechanical and chemical effects that inactivate bacteria and deagglomerate bacterial clusters or flocs (Joyce *et al.*, 2003). The high temperatures produced during cavitation may also be responsible; changes occur momentarily, because only the liquid in the immediate surroundings is heated and therefore only a small number of cells are affected. Ultrasonic waves in water have been shown to form radicals due to homolytic cleavage ($H_2O \rightarrow H^{\cdot} + OH^{\cdot}$). The hydroxy and hydroxyl radicals formed in this reaction are highly reactive and rapidly interact with other radical or chemical species in solution. $H^{\cdot}$ atoms are highly reducing in nature while $OH^{\cdot}$ radicals are highly oxidizing. A common product of this reaction in water is hydrogen peroxide. However, scientists agree that the mechanism of microbial

Table 21.1 List of high-power ultrasound applications and characteristics

Application	Mechanism/effect	Advantages
Crystallization	Cavitation induces the formation of nucleation active sites and creates smaller crystals with modified properties	Decreased crystallization induction times, increased nucleation rate and modified polymorphic crystallization, microstructure, texture and melting behaviour enhanced the heat and mass transfer characteristics
Defoaming	Dissolved gas/oxygen moves towards cavitation bubbles, which grow in size by coalescence and rise to release the entrapped gas to the environment	Increased reduction or elimination of antifoam chemicals and reduced wastage in bottling lines
Drying	Uniform heat transfer	Less time and improved heat transfer
Emulsification	High shear microstreaming	Cost-effective emulsion formation, facilitates the formation of small (40 nm) nanoemulsions and decreased amount of surfactants
Homogenization	Collapse of cavitation forms high energy microjets near interfaces	More stable droplets
Enzyme and microbial inactivation	Increased heat transfer and high shear, physical, mechanical and chemical effects of acoustic cavitation, and direct cavitational damage to microbial cell membranes	Enzyme inactivation adjunct at lower temperatures for improved quality attributes increased the inactivation rate and accelerated enzymatic hydrolysis

(*continued overleaf*)

Table 21.1 (*continued*)

Application	Mechanism/effect	Advantages
Extraction	Increased mass transfer of solvent, release of plant cell material (cavitational dislodgement), cavitation generates high shear forces and microbubbles that enhances surface erosion, fragmentation and mass transfer	Increased extraction efficiency, yield in generally recognized as safe (GRAS) solvent, aqueous or supercritical systems, high yield of extracted materials and fast rate of extraction, minimum effect on extractable materials and enhancement of the extraction of heat-sensitive bioactive and food components at lower processing temperatures
Extrusion	Mechanical vibration and reduced friction	Increased throughput
Fermentation	Improved substrate transfer and stimulation of living tissue	Increasing production of metabolites and acceleration of fermentation processes
Filtration	Disturbance of the boundary layer	Increased flux rates, reduced fouling, less time and improved filtration process
Heat transfer	Improved heat transfer through acoustic streaming and cavitation	Acceleration of heating, cooling and drying of products at low temperatures
Separation	Agglomeration of components at pressure nodal points	Adjunct for use in nonchemical separation procedures
Viscosity alteration	Reversible and nonreversible structural modification via vibrational and high-shear microstreaming	Nonchemical modification for improved processing traits, reduced additives and differentiated functionality

inactivation is mainly due to thinning of cell membranes, localized heating and production of free radicals (Butz and Tauscher, 2002). The effects, however, are not severe enough for sufficient destruction of microorganisms when using ultrasound (US – ultrasonication) alone at a lower temperature. To improve the microbial inactivation, ultrasound is combined with other treatments such as pressure (manosonication), heat (thermosonication), both pressure and heat (manothermosonication) and antimicrobials (Piyasena, Mohareb and McKellar, 2003; Raso and Barbosa-Canovas, 2003). The details of these processes are given in the following subsections.

Thermosonication (TS) In this method, the product is subjected to ultrasound and moderate heat simultaneously. This technique shows the same inactivation level compared to the treatment without ultrasound at high temperatures.

Manosonication (MS) It provides the possibility to inactivate enzymes and/or microorganisms by combining ultrasound with a moderate pressure of 100–300 kPa at low temperatures.

Manothermosonication (MTS) It combines the ultrasound with moderate temperature and moderate pressure in order to inactivate enzymes and/or microorganisms. The ultrasound generates the cavitation or bubble implosion in the media. The simultaneous pressure treatment maximizes the intensity of the explosion, which increases the level of inactivation. Compared to high-power ultrasound (HPU) alone, these treatments are more energy-efficient and effective in inactivating the microorganisms.

The ultrasound has a low lethal effect at ambient temperature and pressure, where the lethality levels are increased with increasing static pressure and/or temperature. Raso *et al.* (1998) have suggested an equation for predicting D values when using manothermosonication. Assuming that the heat and ultrasonic waves affect the medium independent of one another, the D value of manothermosonication can be predicted using Equation (21.3) to give

$$D_{MTS} = (D_T \times D_{MS}) \; / \; (D_T + D_{MS}) \tag{21.3}$$

where D_{MTS} is the decimal reduction time of manothermosonication (min), D_T is the decimal reduction of thermal treatment (min) and D_{MS} is the decimal reduction of manosonication treatment (min).

Herceg, Juraga, Sobota and Režek Jambrak (2012) have suggested an equation for predicting D values when using thermosonication, assuming that ultrasound and temperature act independently and that heat and ultrasound inactivation of microorganisms follow the first-order kinetics. The logarithmic order of inactivation of microorganisms can be expressed by the following

equation (the developed model is based on the model of Raso *et al.*, 1998):

$$\frac{N_t^{TS}}{N_0} = \frac{N_t^S}{N_0}\frac{N_t^T}{N_0} \tag{21.4a}$$

$$\log\frac{N_t^{TS}}{N_0} = \log\left(\frac{N_t^S}{N_0}\,\frac{N_t^T}{N_0}\right) \tag{21.4b}$$

$$\log\frac{N_t^{TS}}{N_0} = \log\frac{N_t^S}{N_0} + \log\frac{N_t^T}{N_0} \tag{21.4c}$$

$$-\frac{t}{D_{TS}} = -\frac{t}{D_S} - \frac{t}{D_T} \tag{21.4d}$$

$$\frac{1}{D_{TS}} = \frac{1}{D_S} + \frac{1}{D_T} \tag{21.4e}$$

$$\frac{1}{D_{TS}} = \frac{D_T + D_S}{D_S\,D_T} \tag{21.4f}$$

$$D_{TS} = \frac{D_T\,D_S}{D_T + D_S} \tag{21.4g}$$

where N_0 is the number of microorganisms before treatment, N_t^T is the number of microorganisms after time t and thermal processing, N_t^S is the number of microorganisms after time t and ultrasound treatment, N_t^{TS} is the number of microorganisms after time t and thermal processing and ultrasound treatment, D_T is the decimal reduction time during thermal processing, D_S is the decimal reduction time during ultrasound treatment and D_{TS} is the decimal reduction time during thermal processing and ultrasound treatment.

There are often significant deviations in the observed linearity of microbial inactivation by applying new methods of food processing. When survival curves are nonlinear, the D value is usually determined by considering the linear portion of the survival curve. Over the years, a number of models have been proposed to describe these nonlinear survival curves, such as the Cerf, modified Gompertz, log-logistic, Baranyi and Weibull models (Ugarte-Romero *et al.*, 2006; Hassani *et al.*, 2005). Among them, the Weibull model is gaining popularity due to its simplicity and flexibility. This model assumes that cells and spores in the population have different resistances and the survival curve is just a form of the cumulative distribution of lethal factors:

$$\log_{10}\frac{N}{N_0} = -bt^n \tag{21.5}$$

Here, b and n are the scale and shape factors.

Although the Weibull model describes nonlinear survival curves better than the linear model, with one more parameter the Weibull model is intrinsically more complex. The concept of D and z values is no longer valid in these nonlinear cases. It is proposed that the Weibull distribution parameters b and n are affected by external conditions, such as temperature, pH, pressure, etc. (Peleg and Cole, 2000). Mattick *et al.* (2001) have found that these two parameters are temperature dependent. Factors affecting the effectiveness of microbial inactivation are the amplitude of ultrasound waves, exposure or contact time, volume of food processed, composition of food and treatment temperature (Chemat, Huma and Kamran Khan, 2011). The effectiveness of ultrasound on the inactivation of microorganisms is also affected by type, shape and size of the microorganisms. Herceg *et al.* (2012) have investigated the effect of the combined ultrasound and heat treatments against ultrasound treatment alone on the inactivation of *Escherichia coli* and *Staphylococcus aureus* in milk. The parameters that seem to substantially affect the inactivation are the amplitude of the ultrasonic waves, the exposure/contact time with the microorganisms and the temperature of the treatment. Gram-negative bacteria (*E. coli*) are more susceptible to the ultrasonic treatment than the gram-positive ones (*S. aureus*). Gram-positive bacteria are known to be more resistant than gram-negative ones, possibly because of their thicker cell wall, which provides them with better protection against ultrasound effects. However, it is probable that there is a significant impact on the size of the cells to inactivate microorganisms (Table 21.2). Larger cells are more sensitive than the small ones. This is probably due to their larger surface area. Concerning the shape of the microorganisms, cocci are more resistant than bacilli due to the relationship of cell surface area and volume. Also, the resistance of different species to ultrasound differs widely. Sporulated microorganisms are more resistant than vegetative ones and fungi are more resistant in general than vegetative bacteria. The commonly applied frequency ultrasound is 20 kHz for microbial inactivation. The resistance to ultrasound treatment at this frequency of spores and gram-positive and coccal cells are higher than vegetative, gram-negative and rod-shaped

Table 21.2 Structure of cell wall and size of cell, which affects the sensitivity of bacteria

Type of microorganism	Structure of cell wall	Cell size (μm)	Shape
Escherichia coli	Gram-negative	1.5 × 6	Straight rods
Staphylococcus aureus	Gram-positive	0.8 × 1	Irregular coccoid
Salmonella typhimurium	Gram-negative	1.5 × 5	Straight rods
Lysteria monocytogenes	Gram-positive	0.5 × 2	Short rods
Bacillus subtilis	Gram-positive	0.5 – 2.5 × 1.5 – 10	Rods with rounded or squared ends

bacteria (Feng, Yang and Hielscher, 2008). In addition, it also varies among different strains. In terms of microbial resistance, more research is necessary about the potential enhancement of the ultrasound in combination with other preservation factors, among them antimicrobials, especially for fungi.

21.3.2 Ultrasound as pre-treatment

Conventional hot-air drying is a very energy- and cost-intensive process. Drying is a simultaneous operation of heat and mass exchange that is followed by phase changes. Application of different pretreatments, like osmotic dehydration, ultrasound and ultrasound-assisted osmotic dehydration has shown different effects on fruits and vegetables. Power ultrasound also improves heat and mass transfer phenomena in drying processes (Cárcel *et al.*, 2011). Gallego-Juárez *et al.* (2007) have utilized an air-borne power ultrasound generator and a procedure in which ultrasonic vibrations are applied in direct contact with the product to be dried and under a certain static pressure. They have designed a prototype based on a high-power rectangular plate transducer, working at a frequency of 20 kHz with a power capacity of about 100 W. The limiting feature for the use of ultrasound in air is the severe absorption, which rapidly reduces the amplitude of the field as it propagates away from the source. Another approach is to use ultrasound as a pre-treatment method prior to drying of mushrooms, brussels sprouts and cauliflower (Jambrak *et al.*, 2007). Pre-treatment with a 20 kHz probe and 40 kHz bath for 3 and 10 min has been compared with blanched (80 °C/3 min) and untreated samples. The drying time after ultrasound treatment is reduced for all samples when compared to an untreated sample.

21.3.3 Ultrasound in filtration

Separation of solids from liquids is an important procedure for the production of solid-free liquid or to produce solid isolated from original liquor. Deposition of solid materials on the surface of a filtration membrane is one of the main problems. The application of ultrasonic energy can increase the flux by breaking the concentration polarization and cake layer at the membrane surface without affecting the intrinsic permeability of the membrane. Ultrasonic-assisted filtration has been successfully employed to enhance the filtration of industrial wastewater that is generally considered difficult to process and to improve the cleaning of fouled membranes (Chemat, Huma and Kamran Khan, 2011). There are two specific effects of ultrasonic irradiation that can be harnessed to improve the filtration technique: (1) sonication causes agglomeration of fine particles (i.e. more rapid filtration) and (2) it supplies sufficient vibrational energy to the system to keep the particles partly suspended and therefore leaves more free ‘channels’ for solvent elution.

21.3.4 Ultrasound-assisted extraction

Ultrasound-assisted extraction (UAE) is an emerging technology that can accelerate heat and mass transfer and has been successively used in the field of extraction. Ultrasound waves, after interaction with plant material, alter its physical and chemical properties. The cavitational effect facilitates the release of extractable compounds and enhances the mass transport by disrupting the plant cell walls. The process of UAE can be optimized, by means of an experimental design, through the operating conditions (solvent concentration, extraction temperature and extraction time) (Chemat, Huma and Kmran Khan, 2011). UAE is as effective as any other high-temperature long-time extraction process because it can greatly decrease the extraction time. The efficiency of UAE can be explained by the fact that sonication simultaneously enhances the hydration and fragmentation process while facilitating the mass transfer of solutes to the extraction solvent (Soria and Villamiel, 2010).

21.3.5 Changes in viscosity and texture, polymerization and depolymerization

Krešić *et al.* (2008) have studied the effect of high-power ultrasound treatment of whey protein isolate and concentrate samples on their rheological properties after ultrasound treatment. Ultrasound causes a significant increase in apparent viscosity and significant changes in flow behaviour indices accompanied by an increase in a consistency coefficient compared to control samples. It is due to changes in the binding capacity for water and altered protein structure after ultrasound treatment. Krešić *et al.* (2011) have also studied biphasic systems with whey proteins and guar gum aqueous dispersions, treated with ultrasound of 30 kHz for 5 and 10 min. Ultrasound treatment shows a significant influence on rheological properties through protein denaturation caused by cavitation and microstreaming effects. Režek Jambrak *et al.* (2011) have studied the effect of ultrasound (30 kHz) on rheological properties of model systems prepared with whey protein isolates or concentrates with or without sucrose or milk powder. There have been changes in the consistency coefficient (k), but no significant changes occur in the flow behaviour indices (n). Režek Jambrak *et al.* (2009) have studied the impact of ultrasound treatment on soy protein isolate and concentrate dispersions. Ultrasound treatment causes statistically significant changes in flow behaviour indices (n) and consistency coefficients (k).

Rheological characteristics of corn starch model systems have been studied by Režek Jambrak *et al.* (2010a) and Ljubić Herceg *et al.* (2010); a statistically significant decrease in consistency coefficient (k) has been noticed as compared to the untreated sample.

21.3.6 Oxidation reaction produced by sonication

Ultrasound in an aqueous medium produces highly reactive species such as OH radicals, H_2O_2 and ozone, which are strong oxidizing agents. These radicals are capable of initiating and enhancing oxidation and reduction reactions. Under the action of cavitation, water decomposes into free radicals. The rate of sonochemical oxidation in an aqueous solution is about three times faster in an air atmosphere compared to an argon environment. The H^+, OH^- and H_2O_2 produced by ultrasound in an aqueous solution are responsible for the oxidation reaction. The dose–response relation is linear for different intensities of ultrasound. A 20 kHz frequency ultrasound produces 14 times more hydroxyl radicals than those produced by 3.5 MHz. Chemat *et al.* (2004) have studied the effect of ultrasound treatment during food emulsification and processing of sunflower, olive and soybean oils. They found a significant negative change in their composition due to the oxidation produced during the ultrasound treatment.

21.3.7 Ultrasound in enhancing fermentation

Production of ethanol from lactose by fermentation with the yeast *Kluyveromyces marxianus* (ATCC 46537) under various sonication regimens has been carried out with a low-intensity sonication. All sonication treatments improve ethanol production relative to the control (no sonication) (Sulaiman *et al.*, 2011). Low-power ultrasonic (20–30 kHz) has been used to enhance the ethanol fermentation from molasses, and the results indicate that ultrasonic power enhances the ethanol production rate by reducing the fermentation time by 6–9 h compared to that using a conventional bioreactor (Klomklieng and Prateepasen, 2011).

21.3.8 Ultrasound-assisted freeze drying

High-intensity ultrasound can be applied to accelerate and improve the efficiency of the mass and heat transfer process at a low temperature between a solid or semi-solid matrix and a gaseous medium. Garcia Perez *et al.* (2012) have studied the application of power ultrasound during atmospheric freeze drying to accelerate the drying process by introducing mechanical energy in the drying chamber. Carrots are subjected to atmospheric power ultrasound-assisted freeze drying (USAFD) at an acoustic power of 20.5 kW/m^3. An increase in the drying rate and a reduction of drying time of 75% have been observed. Schössler, Jäger and Knorr (2012) have freeze dried red bell pepper cubes on a stainless steel screen (used as the sound transmitting surface). The ultrasound treatment reduces the drying time required to reach a final moisture content of 10% (dry basis) by 11.5%. Bantle and Eikevik (2011) have also investigated the influence of the drying

temperature, drying time and ultrasonic power for atmospheric freeze drying in the presence of an airborne ultrasonic field. An accelerated effective diffusion of up to 14.8% has been obtained for atmospheric freeze drying with a fluid bed.

21.3.9 Ultrasound in modification of functional properties

Režek Jambrak *et al.* (2008) have studied the effect of sonication on whey proteins in order to improve their functional properties. Ultrasound (20 kHz) affects the function of whey proteins like solubility and foaming ability by sample exposure at high temperatures caused by sonication. Another study by Režek Jambrak *et al.* (2010b) on α -lactalbumin shows that the solubility increases significantly for all samples at 20 kHz treatment. Foam capacities and foam stabilities are improved after ultrasound treatments for both 20 and 40 kHz treatments. Jackson *et al.* (1988) have used ultrasound to dissolve corn and sorghum starch granules after heating for the analysis of molecular structures. They reported that ultrasonic vibrations disrupt swollen granules, thereby releasing amylose and amylopectin from the granules and resulting in an increase in the water solubility of starch. Chung *et al.* (2002) have treated mung bean, potato and rice starches with ultrasound after heating; the changes in starch properties have been related to the disruption of swollen granules. Ljubić Herceg *et al.* (2010) have studied the effect of ultrasound (24 kHz) on corn starch dispersions; the disruption of starch granules by cavitational forces made the granules more permeable to water. Jambrak *et al.* (2007) examined the effect of high-power ultrasound of 24 kHz frequency on selected physical properties of corn starch. The results indicate the distortion of starch in the crystalline region. Zhang *et al.* (2011) have studied the effect of ultrasound on the foaming and emulsifying properties of wheat gluten. The foam capacity and foam stability of ultrasound-treated wheat gluten proteins gradually increase as the treatment power has been increased and are more pronounced at the 100% power level.

21.4 Conclusions

Food processing employing ultrasound technology has been the object of research by food scientists in the last ten years. This technology has proved to be a promising technique in laboratory and industrial applications. It has several advantages in technical, economical and energy aspects. Ultrasound has been shown to serve as a technique that can be used in many traditional food processing operations and processes, and innovative applications and designs of equipment are available. However, there are some disadvantages of using this technology as the production of free radicals is possible that

can damage the product. Using ultrasound in a proper way by selecting appropriate amplitude, power, intensity, temperature, pressure and time is crucial to obtain the desired product.

References

Ashokkumar, M. and Kentish, S. (2011) The physical and chemical effects of ultrasound. In: *Ultrasound Technologies for Food and Bioprocessing* (Feng, H., Barobosa-Cànovas, G.V. and Weiss, J., eds). Springer, New York, USA, pp. 1–105.

Bantle, M. and Eikevik, T.M. (2011) Parametric study of high-intensity ultrasound in the atmospheric freeze drying of peas. *Drying Technology*, **29**, 1230–1239.

Bermudez-Aguirre, D., Mobbs, T. and Barbosa-Canovas, G.V. (2011) Ultrasound applications in food processing. In: *Ultrasound Technologies for Food and Bioprocessing* (Feng, H., Barobosa-Cànovas, G.V. and Weiss, J., eds). Springer, New York, USA, pp. 65–105.

Butz, P. and Tauscher, B. (2002) Emerging technologies: chemical aspects. *Food Research International*, **35**, 279–284.

Cárcel, J.A., Garcia Perez, J.V., Riera, E. and Mulet, A. (2011) Improvement of convective drying of carrot by applying power ultrasound – influence of mass load density. *Drying Technology*, **29**, 174–182.

Chandrapala, J., Oliver, C., Kentish, S. and Ashokkumar, M. (2012) Ultrasonics in food processing. *Ultrasonics Sonochemistry*, **19**, 975–983.

Chemat, F., Huma, Z. and Kamran Khan, M. (2011) Applications of ultrasound in food technology: processing, preservation and extraction. *Ultrasonics Sonochemistry*, **18**, 813–835.

Chemat, F., Grondin, I., Shum Cheong Sing, A. and Smadja, J. (2004) Deterioration of edible oils during food processing by ultrasound. *Ultrasonics Sonochemistry*, **11**, 13–15.

Chung, K.M., Moon, T.W., Kim, H., and Chun, J.K. (2002) Physicochemical properties of sonicated mung bean, potato, and rice starches. *Cereal Chemistry*, **79**, 631–633.

Feng, H., Yang, W. and Hielscher, T. (2008) Power ultrasound. *Food Science and Technology International*, **14**(5), 433–436.

Gallego-Juárez, J.A., Riera, E., Blanco, S.F., Rodríguez-Corral, G., Acosta-Aparicio, V.M. and Blanco, A. (2007) Application of high-power ultrasound for dehydration of vegetables: processes and devices. *Drying Technology*, **25**, 1893–1901.

Garcia Perez, J.V., Santacatalina, J.V., Carcel, J.A., Simal, S. and Mulet, A. (2012) Atmospheric freeze drying assisted by power ultrasound. In: *International Symposium on Ultrasound in the Control of Industrial Processes*, UCIP, 2012, Madrid, Spain.

Gogate, P.R. and Kabadi, A.M. (2009) A review of applications of cavitation in biochemical engineering/biotechnology. *Biochemical Engineering Journal*, **44**, 60–72.

Hassani, M., Álvarez, I., Raso, J., Condón, S. and Pagán, R. (2005) Comparing predicting models for heat inactivation of *Listeria monocytogenes* and *Pseudomonas aeruginosa* at different pH. *International Journal of Food Microbiology*, **100**, 213–222.

Herceg, Z., Juraga, E., Sobota, Š.B. and Režek Jambrak, A. (2012) Inactivation of mesophilic bacteria in milk by means of high intensity ultrasound using response surface methodology. *Czech Journal of Food Science*, **30**, 108–117.

Herceg, Z., Režek Jambrak, A., Lelas, V. and Mededovic, T.S. (2012) The effect of high intensity ultrasound treatment on the amount of *Staphylococcus aureus* and *Escherichia coli* in milk. *Food Technology and Biotechnology*, **50**, 46–52.

Herceg, Z., Markov, K., Šalamon, B.S., Režek Jambrak, A., Vukušić, T. and Kaliterna, J. (2013) Effect of high intensity ultrasound treatment on growth of food spoilage bacteria. *Food Technology and Biotechnology*, **51**(3), 352–359.

Hoffmann, M.R., Hua, I. and Höchemer, I.H.R. (1996) Application of ultrasonic irradiation for the degradation of chemical contaminants in water. *Ultrasonics Sonochemistry*, **3**, 163–172.

Jackson, D.S., Choto-Owen, C., Waniska, R.D. and Rooney, L.W. (1988) Characterization of starch cooked in alkali by aqueous high-performance size exclusion chromatography. *Cereal Chemistry*, **65**, 493–496.

Jambrak, A.R., Mason, T.J., Paniwnyk, L. and Lelas, V. (2007) Ultrasonic effect on pH, electric conductivity, and tissue surface of button mushrooms, Brussels sprouts and cauliflower. *Czech Journal of Food Sciences*, **25**(2), 90–100.

Jambrak, A.R., Mason, T.J., Paniwnyk, L. and Lelas, V. (2007) Accelerated drying of button mushrooms, Brussels sprouts and cauliflower by applying power ultrasound and its rehydration properties. *Journal of Food Engineering*, **81**, 88–97.

Joyce, E., Phull, S.S., Lorimer, J.P. and Mason, J.T. (2003) The development and evaluation of ultrasound for the treatment of bacterial suspensions. A study of frequency, power and sonication time on cultured *Bacillus* species. *Ultrasonic Sonochemistry*, **10**, 315–318.

Juraga, E., Sobota, Š.B., Herceg, Z. and Režek Jambrak, A. (2011) Application of high intensity ultrasound treatment on *Enterobacteriae* count in milk. *Mljekarstvo*, **61**, 131–140.

Klomklieng, W. and Prateepasen, A. (2011) Using low-power ultrasonic for enhancing *Saccharomyces cerevisiae* M30 productivity for ethanol produced from molasses. In: *International Conference on Food Engineering and Biotechnology*, IPCBEE, vol. 9, Singapore, pp. 234–240.

Krešić, G., Lelas, V., Režek Jambrak, A., Herceg, Z. and Rimac Brnčić, S. (2008) Influences of novel food processing technologies on the rheological and thermophysical properties of whey proteins. *Journal of Food Engineering*, **87**, 64–73.

Krešić, G., Režek Jambrak, A., Lelas, V. and Herceg, Z. (2011) Influence of innovative technologies on rheological and thermophysical properties of whey proteins and guar gum model systems. *Mljekarstvo*, **61**(1), 64–78.

Leonelli, C. and Mason, T.J. (2010) Microwave and ultrasonic processing: now a realistic option for industry. *Chemical Engineering and Processing*, **49**(9), 885–900.

Ljubić Herceg, I., Režek Jambrak, A., Šubarić, D., Brnčić, M., Rimac Brnčić, S., Badanjak, M., Tripalo, B., Ježek, D., Novotni, D. and Herceg, Z. (2010) Texture and pasting properties of ultrasonically treated corn starch. *Czech Journal of Food Sciences*, **28**(2), 83–93.

Mason, T. (1998) Power ultrasound in food processing – the way forward. In: *Ultrasound in Food Processing* (Mason, T.J. and Povey, M.J.W., eds). Blackie Academic and Professional, London, England, pp. 105–125.

Mason, T.J. and Cordmas, E. (1996) Ultrasonic intensification of chemical processing and related operations – a review. *Transactions of the Institute of Chemical Engineers*, **74**(A), 511–516.

Mattick, K.L., Legan, J.D., Humphrey, T.J. and Peleg, M. (2001) Calculating *Salmonella* inactivation in non-isothermal heat treatments from isothermal non-linear survival curves. *Journal of Food Protection*, **64**, 606–613.

O'Brien Jr, W.D. (2007) Ultrasound–biophysics mechanisms. *Progress in Biophysics and Molecular Biology*, **93**, 212–255.

Patist, A. and Bates, D. (2008) Ultrasonic innovations in the food industry: from the laboratory to commercial production. *Innovative Food Science and Emerging Technologies*, **9**, 147–154.

Peleg, M. and Cole, M.B. (2000) Estimating the survival of *Clostridium botulinum* spores during heat treatments. *Journal of Food Protection*, **63**, 190–195.

Piyasena, P., Mohareb, E. and McKellar, R.C. (2003) Inactivation of microbes using ultrasound: a review. *International Journal of Food Microbiology*, **87**, 207–216.

Raso, J. and Barbosa-Canovas, G.V. (2003) Nonthermal preservation of foods using combined processing techniques. *Critical Reviews of Food Science and Nutrition*, **43**, 265–285.

Raso, J., Pagan, R., Condon, S. and Sala, F.J. (1998) Influence of temperature on the lethality of ultrasound. *Applied Environmental Microbiology*, **64**, 465–471.

Režek Jambrak, A., Mason, T.J., Lelas, V., Herceg, Z. and Ljubić Herceg, I. (2008) Effect of ultrasound treatment on solubility and foaming properties of whey protein suspensions. *Journal of Food Engineering*, **86**, 281–287.

Režek Jambrak, A., Lelas, V., Mason, T.J., Krešić, G. and Badanjak, M. (2009) Physical properties of ultrasound treated soy proteins. *Journal of Food Engineering*, **93**, 386–393.

Režek Jambrak, A., Herceg, Z., Šubarić, D., Babić, J., Brnčić, M., Rimac Brnčić, S., Bosiljkov, T., Čvek, D., Tripalo, B. and Gelo, J. (2010a) Ultrasound effect on physical properties of corn starch. *Carbohydrate Polymers*, **79**, 91–100.

Režek Jambrak, A., Mason, T.J., Lelas, V. and Krešić, G. (2010b) Ultrasonic effect on physicochemical and functional properties of a-lactalbumin. *LWT - Food Science and Technology*, **43**, 254–262.

Režek Jambrak, A., Lelas, V., Krešić, G., Badanjak, M., Rimac Brnčić, S., Herceg, Z., Batur, V. and Grčić, I. (2011) Rheological, functional and thermo-physical properties of ultrasound treated whey proteins with addition of sucrose or milk powder. *Mljekarstvo*, **61**, 79–91.

Santos, H.M., Lodeiro, C. and Capelo, J.L. (2009) Ultrasonic energy as a tool for sample treatment for the analysis of elements and elemental speciation. In: *Ultrasound in Chemistry. Analytical Applications* (Capelo-Martínez, J.L., ed.). Wiley VCH Verlag GmbH&Co. KGaA, Weinheim, Germany, pp. 17–50.

Schössler, K., Jäger, H. and Knorr, D. (2012) Novel contact ultrasound system for the accelerated freeze-drying of vegetables. *Innovative Food Science and Emerging Technologies*, **16**, 113–120.

Soria, A.C. and Villamiel, M. (2010) Effect of ultrasound on the technological properties and bioactivity of foods: a review. *Trends in Food Science and Technology*, **21**, 323–331.

Starritt, H.C., Duck, F.A. and Humphrey, V.F. (1989) An experimental investigation of streaming in pulsed diagnostic ultrasound beams. *Ultrasound Medicine and Biology*, **15**, 363–373.

Sulaiman, A.Z., Ajit, A., Yunus, R.M. and Chisti, Y. (2011) Ultrasound-assisted fermentation enhances bioethanol productivity. *Biochemical Engineering Journal*, **54**, 141–150.

Ugarte-Romero, E., Feng, H., Scott, E.M., Cadwallader, K.R. and Robinson, S.J. (2006) Inactivation of *Escherichia coli* with power ultrasound in apple cider. *Journal of Food Science*, **71**(2), 102–108.

Villamiel, M. and de Jong, P. (2000) Inactivation of *Pseudomonas fluorescens* and *Streptococcus thermophilus* in trypticase soy broth and total bacteria in milk by continuous-flow ultrasonic treatment and conventional heating. *Journal of Food Engineering*, **45**, 171–179.

Zhang, H., Claver, I.P., Zhu, K.X. and Zhou, H. (2011) The effect of ultrasound on the functional properties of wheat gluten. *Molecules*, **16**, 4231–4240.

22
Membrane Processing

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22.1 Introduction

Membrane operations have become major tools in the food processing industries over the last 30 years. With a market volume of 800–850 million euros/year, the food industry represents the second biggest worldwide industrial market for membranes after water and wastewater treatment (Lipinzki, 2010). The growth in this market is around 7.5% per year (Daufin *et al.*, 2001).

The basic properties of membrane operations make them ideal for the treatment of both food products and by-products; they are generally athermal and do not involve phase changes or chemical additives; they are simple in concept and operation, modular and easy to scale-up; furthermore, they are characterized by a low energy consumption, permitting a rational utilization of raw materials and recovery and reuse of by-products.

Pressure-driven membrane operations, such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) are well-established technologies in the dairy industry, followed by beverages (wine, beer, fruit juices, etc.), egg products and process water. New membrane processes, such as pervaporation (PV), electrodialysis (ED), membrane bioreactors (MBRs) and membrane contactors (MCs), offer interesting perspectives in the revision of traditional flow sheets in food processing. Furthermore, the combination of different membrane operations within the logic of process intensification strategy offers new and many more opportunities in terms of competitiveness, improvement of quality, process or product novelty and environmental friendliness (Drioli and Romano, 2001).

The present chapter gives an outlook of both well-established and emerging membrane applications in the food industry.

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22.2 Terminology and general considerations

A membrane is defined as a structure with lateral dimensions much greater than its thickness through which mass transfer can occur under a variety of driving forces, such as gradient of concentration, pressure, temperature, electric potential, etc (Koros, Ma and Shimidzu, 1996). It is in general a solid or a liquid film that might ideally represent an interface between two phases.

Membranes can be classified according to their nature, geometry and separation regime (Khulbe, Feng and Matsuura, 2008). Most commercial membranes are formed from organic polymers or from inorganic materials (ceramic, metallic, carbon, etc). According to their geometry, membranes can be subdivided into tubular, hollow fibre, spiral wound and flat sheet (Mallevialle, Odendaal and Wiesner, 1988). Tubular membranes offer no dead space, do not get blocked easily and are easy to clean. However, as the tube diameter increases, they occupy a larger space, have a higher hold-up volume and require high pumping costs. Tube diameters are typically in the range of 10–25 mm. Hollow fibre membranes, with diameters of 0.001–1.2 mm, offer high packing density and can withstand relatively high pressures. They may not perform well with viscous feeds and solutions containing particulate matter. Consequently, an extensive pre-treatment of the solution is needed in order to remove particles, macromolecules or other materials that can precipitate at the membrane surface. Due to the self-supporting nature of the membrane, hollow fibre systems can be submitted to a physical process called backflushing in which the permeate flow is reversed to dislodge the fouling material from the membrane surface, allowing a more easy control of concentration polarization and fouling phenomena. Spiral wound membranes consist of a sandwich of flat-sheet membranes, spacers and porous permeate flow material wrapped around a central permeate collecting tube. They offer advantages such as compactness, good membrane surface/volume and low capital/operating cost ratios. Nevertheless, they cannot be mechanically cleaned and a feed pre-treatment is required. Commercial systems are about 1 metre long with diameters between 10 and 60 cm. Membrane areas can be in the range 3 – 60 m^2.

Flat-sheet membranes offer moderate membrane surface/volume ratios. However, the low packing density restricts their use to clear feed streams containing only fine suspended solids; in addition, these systems are quite expensive and the exchange of membranes is labour intensive.

Membranes can be also classified on the basis of the separation mechanism determined by specific properties of the components (Mulder, 1991): (1) separation based on molecules/membrane surface interactions (e.g. multilayer diffusion) and/or the difference between the average pore diameter and the average free path of fluid molecules (e.g. Knudsen mechanism), (2) separation based on the difference of diffusivity and solubility of substances in the

membrane (solution-diffusion mechanism), and (3) separation based on the difference in charge of the species to be separated (electrochemical effect).

Based on these separation mechanisms, membranes can be further classified into *dense, porous* and *ion-exchange membranes*. Finally, membranes can be symmetric or asymmetric if they are homogeneous solid films, microporous media or electrically charged barriers. Symmetric membranes are characterized by an identical structure and transport properties over the whole cross-section. The flux is generally determined by membrane thickness. Asymmetric membranes have a thin (0.1 – 1 μm) dense 'skin' layer supported by a 100 – 200 μm thick macroporous layer. Separation characteristics are determined by the nature of the membrane material or the pore size of the skin layer. A classification of synthetic membranes based on their structure is summarized is Figure 22.1.

Porous membranes consist of a solid matrix with defined pores having diameters ranging between 1 nm and 10 μm. Components are separated mainly thorough a sieving mechanism in which pore diameters and particle size are the determining parameters. Consequently, these membranes are mainly used for separating compounds with different sizes or molecular weights, such as MF, UF or dialysis (Cheryan, 1998). Dense membranes do not present any detectable pores at the limits of electron microscopy (< 5 nm) throughout their thickness. A solution-diffusion mechanism is mainly involved in the separation of compounds having different diffusivities and solubilities in the membrane matrix. Typically these membranes are used to separate compounds with a similar size. Electrically charged membranes (ion-exchange membranes) consist of films carrying fixed positive or negative charges. The main application of ion-exchange membranes is in ED, where the charge density is responsible for the membrane selectivity. However, a partial contribution of the charge density to the membrane selectivity is observed also in UF and NF membranes.

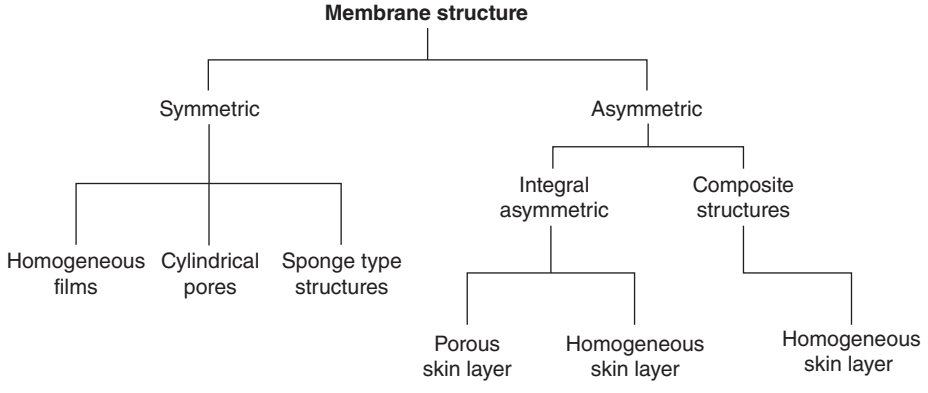

Figure 22.1 Classification of synthetic membranes on the basis of their structure

22.3 Pressure-driven membrane operations

Pressure-driven membrane processes are based on the use of a permselective barrier through which fluids and solutes are selectively transported when a hydrostatic pressure is applied. As a result, the feed solution is converted into a permeate containing all components that have permeated the membrane and a retentate containing all compounds rejected by the membrane. The separation is based mainly on molecular size, but to a lesser extent on shape and charge.

Microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) are typical pressure-driven membrane operations, which differ in the size of particles to be separated. The filtration capability of these processes is shown in Figure 22.2. MF is used to separate particles with diameters of 0.1–10 μm from a solvent or other low molecular weight compounds. These particles are generally larger than those separated by UF and RO. Consequently, the osmotic pressure for MF is negligible and hydrostatic pressure differences used in MF are relatively small (in the range of 0.5–4 bar). UF is a membrane process similar to MF in the mode of separation, based on the use of asymmetric membranes with pore sizes in the skin layer of 2–10 nm. Typically dissolved molecules or small particles not larger than 0.1 μm in diameter are retained. A UF membrane is typically characterized by its molecular weight cut-off (MWCO), defined as the equivalent molecular weight of the smallest species that exhibit 90% rejection. The MWCO of UF membranes is between 10^3 and 10^6 Dalton. Hydrostatic pressures of 2–10 bar are typically used.

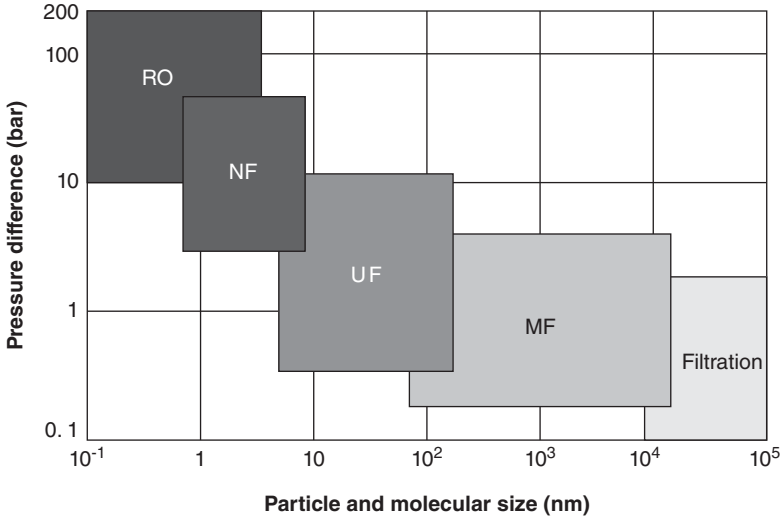

Figure 22.2 Separation capabilities of pressure driven membrane separation processes

In the NF process, components of a fluid are fractionated mainly according to their size and charge. Multivalent ions and uncharged organic molecules with molecular weights between 100 and 1000 Da are typically separated. NF membranes are characterized by a charged surface with pore diameters in the range of 1–3 nm. They operate at lower pressures (generally in the range of 3–30 bar) than RO membranes. RO membranes are typically used to separate low molecular weight compounds from a solvent, usually water. The particle size range for applications of RO is 0.1–1 nm and solutes with molecular weights greater than 300 Da are separated. The hydrostatic pressures applied as the driving force in RO to obtain significant transmembrane flux are of the order of 10–100 bar. The basic properties of pressure-driven membrane operations are summarized in Table 22.1.

All these processes can be operated either in dead-end or in cross-flow configurations. In the dead-end mode, the feed flow is perpendicular to the membrane surface. It is forced through the membrane, which causes the retained particles to accumulate and form a type of cake layer at the membrane surface. The thickness of the cake layer increases with the filtration time. Therefore, the permeation rate decreases by increasing the cake layer thickness. In the cross-flow mode, the fluid to be filtered flows tangentially to the membrane surface and permeates through the membrane due to the imposed transmembrane pressure difference. Unlike dead-end filtration, rejected particles form a cake layer on the membrane surface, which does not build up indefinitely. The cake layer thickness remains relatively thin due to the high shear exerted by the solution flowing tangentially to the membrane surface, keeping away the deposited particles towards the filter exit.

An important consideration for pressure-driven membrane processes is that the separation takes place not in the bulk solution but in a very small region close to the membrane, named the boundary layer, as well as over the membrane itself. In particular, compounds that are partially or

Table 22.1 Basic properties of pressure-driven membrane operations

Process	Driving force	Permeate	Retentate
MF	Pressure difference (0.5–4 bar)	Solvent (water) and dissolved solutes	Suspended solids, fine particulates, some colloids
UF	Pressure difference (2–10 bar)	Solvent (water) and low molecular weight solutes (< 1000 Da)	Macrosolutes and colloids
NF	Pressure difference (3–30 bar)	Solvent (water), low molecular weight solutes, monovalent ions	High molecular weight compounds, multivalent ions
RO	Pressure difference (10–100 bar)	Solvent (water)	All dissolved and suspended solids

completely rejected by the membrane tend to accumulate on the membrane surface during filtration, causing a decline of the transmembrane flux. This phenomenon is referred to as *concentration polarization*. Equally devastating for the performance of pressure-driven membrane processes is *membrane fouling*, which can be considered a long-term flux decline caused by the deposition of retained particles (colloids, macromolecules, dissolved organics, etc.) on to the membrane surface or in the membrane pores. The degree of membrane fouling determines the frequency of cleaning, lifetime of the membrane and the membrane area needed, and consequently it has a significant effect on the cost, design and operation of membrane plants (Strathmann, Giorno and Drioli, 2006; Cheryan, 1998).

Pressure-driven membrane processes are widely used in the dairy industry and most of the industrial developments of membrane technologies in the food industry originate from these applications. The availability of membranes with different MWCOs and pore sizes allows selected milk compounds to be separated from other components, thus allowing the manufacture of products with specific properties. Consolidated applications of MF in milk processing concern the removal of bacteria, the milk standardization, the production of concentrated milk and casein fractionation (Atra *et al.*, 2005).

UF membranes retain proteins, fats, insoluble and bound salts, allowing the permeation of lactose and soluble salts. They can be used to standardize milk proteins, offering a valid solution to natural variations of its composition, and to concentrate milk, with the aim of reducing refrigeration and transportation costs (Ernstrom, Sutherland and Jameson, 1980). The advantages of UF in cheese making are mainly in terms of reduction in the use of enzymes (rennet), increased yields of cheese of 10–30%, reduced volume of milk to handle (with a consequent reduction in the number of cheese-making vats), reduction of produced whey since water and lactose are removed in the process, uniformity in the quality with minimization of seasonal variations and a continuous and automated process (Cheryan, 1998). UF is also largely used for the production of whey protein concentrates (WPCs); the initial protein content of 10–12% (dry basis) can be increased up to 80% with a simultaneous decrease in lactose and some salts. WPCs can be further fractionated into β-lactoglobulin and α -lactoglobulin fractions or be used for the manufacture of macropeptides with pharmacotherapeutic value (Cheang and Zidney, 2003). Whey can be concentrated by RO to reduce transportation costs. In this approach, whey proteins are concentrated initially by UF in a first retentate followed by a NF step aimed at the recovery of lactose. Whey proteins can be also concentrated in a single NF operation (up to 20–22% of dry matter), allowing simultaneous reduction of minerals by 25–50% (Kelly and Kelly, 1995). Other interesting applications of NF in the dairy industry concern the concentration of milk for yogurt manufacture as an alternative to vacuum evaporation and the selective demineralization of yogurt. Finally, the treatment of dairy effluents by NF and

RO offers very interesting opportunities to recover milk proteins and lactose and to recycle the depolluted permeate as rinsing water.

Positive effects on transmembrane fluxes and membrane selectivities have been obtained through the introduction of dynamic or shear-enhanced filtrations. Interesting applications of vibratory membrane systems, such as the so called vibratory shear enhanced process (VSEP), are related to the casein separation from whey proteins by MF (Al-Akoum, Ding and Jaffrin, 2002), the dynamic UF of skim milk (Jaffrin *et al.*, 2004) and the protein concentration by UF for cheese manufacturing (Al-Akoum, Jaffrin and Ding, 2005).

MF and UF membranes can be used successfully to replace the use of fining agents or filter aids in the clarification of fruit juices. They are typically used to separate juices into a fibrous concentrated pulp (retentate) and a clarified fraction free of spoilage microorganisms (permeate). These membranes retain high molecular weight compounds (pectin or proteins) and allow low molecular weight solutes (sucrose, acids, salts, aroma and flavour compounds) to permeate through the membrane. Significant advantages over conventional clarification methodologies are in terms of low energy requirements and costs, possibility of avoiding the use of gelatines, adsorbents and other filtration aids, possibility of a lower temperature of processing, better product quality, possibility of operating in a single step reducing working times, simpler process design, reduction of waste products, possibility of avoiding the use of chemical agents, increased juice yield, easy cleaning and maintenance of the equipment, and possibility of avoiding pasteurization (at 60–65 °C) and sterilization (at about 110 °C) (Girard and Fukumoto, 2000; Fukumoto, Delaquis and Girard, 1998).

The most used configurations for the clarification of fruit juices at the industrial level are tubular (inner diameter 5–10 mm), capillary (1–1.5 mm) and plate-and-frame membrane modules. Vibrating membrane systems are particularly appropriate for juices with a high pulp content since they develop a higher shear rate at the membrane surface, reducing membrane fouling. The use of UF in the clarification of different fruit juices produces an improvement in colour and clarity of the juice through the removal of pectic materials and colloidal particles, which result in a lower viscosity of the filtered juice. Almost all soluble solids are recovered in the permeate. The pH and density are generally not affected by juice filtration (Cardoso de Oliveira, Caleffi Docê and Davanted de Barros, 2012; Cassano, Conidi and Drioli, 2010; Laorko *et al.*, 2010; Cassano, Donato and Drioli, 2007; Cassano, Marchio and Drioli, 2007; Rai *et al.*, 2006; Matta, Moretti and Cabral, 2004; de Bruijn *et al.*, 2003; Vladisavljević, Vukosavljevic and Bukvić, 2003).

Clarified juices can be submitted to a concentration step by using RO. The advantages of RO over traditional evaporation are in low thermal damage to the product, reduction in energy consumption and lower capital investments (Merson, Paredes and Hosaka, 1980), as the process is carried out at low temperatures and it does not involve phase change for water removal.

The main disadvantage of RO is its inability to reach the concentration of standard products produced by evaporation because of high osmotic pressure limitations. Thus, it is used as a pre-concentration technique, which permits concentration values of about 30 °Brix corresponding to osmotic pressures of about 50 bar (Pepper, 1990).

Cross-flow MF and UF operations offer different advantages over the use of diatomaceous-earth filtration in wine clarification in terms of low operating temperatures, reduction of energy consumption, removal of bacteria, cells and spores, reduction of SO_2 addition, reduction of the pectin content, proteins and colloids, and a sterilized and clear product in one single continuous operation (Rayess *et al.*, 2011). UF can be used either before the fermentation (for clarifying the must) or after (for treating the finished wine). UF of the must is employed for the removal of colloids, high molecular weight tannins, polysaccharides, proteins and undesirable microorganisms. After the fermentation process and before storage, UF can be used to improve the stability of the finished wine.

MF membranes with an average pore diameter of 0.2 μm can ensure microbiological limpidity and stability in a single operation, producing wines with low turbidity (less than 1 NTU) (Daufin *et al.*, 2001). MF membranes retain polysaccharides and polyphenols which are considered the main cause of fouling in both MF mineral and organic membranes (Vernhet *et al.*, 1999).

The traditional method of haze removal is based on the use of fining agents, such as bentonite, which also cause a retention of colour and some of flavour compounds. The deposit or haze formation in bottled wines due to protein aggregation during storage is a common defect of commercial wines. The UF process permits the haze-causing proteins to be removed, thus preserving the flavour and colour of the wine. In the brewing process, MF competes with natural sedimentation, centrifugation and filter-press to recover 'green beer' or 'rough beer' from tank bottoms, reducing the beer loss; the permeate recovered by MF can be recycled in the wort or in the maturation vessels (Fillaudeau, Blanpain-Avet and Daufin, 2006). MF produces a permeate of acceptable quality with minimal loss of original gravity, colour and bitterness. It can be operated at low temperatures (close to 0 °C), achieving an economic flux and hygienic beer recovery (Daufin *et al.*, 2001). Additional advantages over conventional methodologies include the elimination of filter aids and associated handling and disposal problems, and substitution of heat pasteurization.

22.4 Electrodialysis

Electrodialysis (ED) is used to separate charged molecules from uncharged ones under the influence of an electric field in a system composed of ion-exchange membranes. These are commonly homogeneous membranes

containing ionic groups ($-NH_3^+$ or $-SO_3^-$). Ion-exchange membranes commonly used in ED are of two different types, such as (1) cation-exchange membranes containing negatively charged groups fixed to the polymer matrix, which therefore allow only the permeation of cations, and (2) anion-exchange membranes containing positively charged groups fixed to the polymer matrix, which allow only the permeation of anions.

A typical ED setup consists of both membrane systems arranged alternately and separated by spacers. Each pair of cation- and anion-exchange membranes is called a cell pair. When an electrical potential is established between two electrodes, cations migrate towards the negatively charged cathode while anions migrate towards the positively charged anode. Cations can easily permeate cation-exchange membranes but will be retained by anion-exchange membranes. On the contrary, anions can easily permeate anion-exchange membranes. Consequently, ions will be accumulated in alternating cells giving the concentrate solution while the other cells, depleted in ions, will form the permeate solution (Figure 22.3). The key advantage of ED over other membrane technologies is represented by its selectivity towards charged molecules without affecting uncharged compounds.

The most important applications of ED in the food processing industries are related to dairy products and beverages. In the dairy industry, a consolidated application concerns the demineralization and acidification of cheese whey. In most commercial plants, ED is carried out as a batch operation on

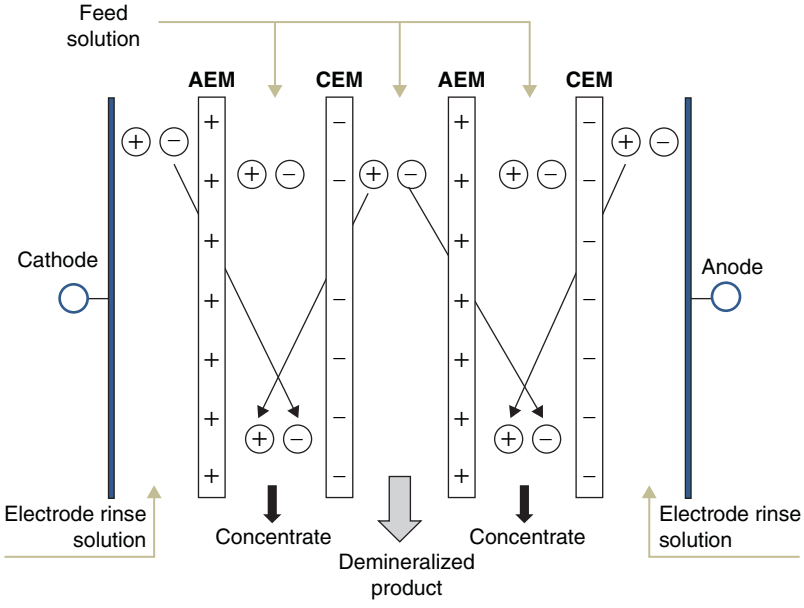

Figure 22.3 Schematic diagram of electrodialysis process (AEM, anion-exchange membrane; CEM, cation-exchange membrane)

pre-concentrated sweet whey; however, acid whey, skim milk, milk and whey UF permeates are also used. The use of concentrated whey permits its ionic concentration to increase, overcoming the limiting factor of ED, represented by the decrease in electrical conductivity of low ionic concentrated solutions. In particular, a two-fold increase of the electrical conductivity has been observed in concentrates obtained from UF permeates by using RO up to a concentration factor of 4 (Lonergan, Fennema and Amundson, 1982). The removal of calcium from skim milk by ED reduces the level of ash and increases the calcium/phosphate ratio, improving the stability of frozen skim milk and concentrated skim milk proteins (Bazinet, 2004).

In the wine industry, ED can be used to extract salts from grape musts and wine to improve their stability. The formation of precipitates can be avoided by reducing potassium concentrations. The amount of ions to be removed is dependent upon the type of wines, grapes and types of vineyards. ED is also commonly used to remove tartrates from wine before bottling. The presence of tartrate in wine causes the formation of precipitates during storage, decreasing the quality of wine.

In the juice and sugar industries, ED is primarily used for the deacidification and demineralization of juices (Vera *et al.*, 2008). The high acidity of some fruit juices (such as orange, grape, pineapple, etc.) make them not acceptable for the market both in single-strength and concentrated forms. Furthermore, in the juice industry the Brix/acid ratio (the ratio of sugars to acid) is specific for each type of juice; for example, an acceptable Brix/acid ratio for grapefruit juice is about 10–11. The sugar addition is commonly used to increase the Brix/acid ratio but this procedure is limited by the high cost of sugar and the blending equipment. The use of ED permits to overcome these limitations. A typical ED equipment for juice deacidification is made of alternate diluate compartments, containing the juice, and concentrate compartments, containing alkaline solutions. In this configuration only anions pass through the membrane, and the global effect is the extraction of anions from the juice and their substitution with OH^- ions from the alkali compartment (Vera *et al.*, 2009).

22.5 Membrane contactors

Membrane contactors (MCs) are devices where separation of compounds is determined by a specific driving force through the membrane from one phase to the other opposite side. These systems allow gas/liquid or liquid/liquid mass transfers without dispersion of one phase within another.

In these processes, the driving force is a concentration and/or pressure difference between the feed and the permeate side, and the separation performance is determined by the distribution coefficient of a component in two phases. Unlike traditional pressure-driven membrane processes, the membrane represents only an interface between two homogenous phases and the

species are transferred from one phase to another by simple diffusion through the membrane pores (Gabelman and Hwang, 1999). MCs have a number of important advantages in contrast with dispersed phase contactors, such as no flooding at a high flow rate, a high and constant interfacial area, high modularity and compatibility, no unloading at a low flow rate, absence of emulsions, easy scale-up and control and the possibility to operate at room temperature. Some limitations are represented by membrane fouling, limited lifetime of the membranes, channelling of fluids and operating pressures depending on critical penetration pressures (Drioli, Curcio and Di Profio, 2005).

The membrane should be properly chosen to enable as much as possible higher values of the mass transfer coefficient. In particular, membranes used in MCs can be hydrophobic and hydrophilic. Typical hydrophobic polymers are polypropylene (PP), polyethylene (PE), polyvinylidene difluoride (PVDF), perfluoropolymers (such as hyflon) and polytetrafluoroehylene (PTFE). They can be wetted by non-polar liquids while polar phases cannot enter into membrane pores. The critical penetration pressure should not be exceeded in order to prevent the penetration of the polar phase into the pores with a consequent loss of hydrophobicity. For a specific material, the critical penetration pressure (ΔP) is given by the Laplace's equation:

$$\Delta P = 2\gamma \frac{\cos\theta}{r} \tag{22.1}$$

where γ is the surface tension of the liquid, θ is the contact angle between the liquid and the membrane and r is the pore radius. According to Equation (22.1), the maintenance of the hydrophobicity is guaranteed by large pore sizes by lowering the penetration pressure.

Hydrophilic membranes are wetted by polar liquids while the non-polar phase remains blocked at the pore mouth. The interface, formed at the pore mouth of the non-polar phase side, is maintained if the critical penetration pressure is not exceeded (Drioli, Criscuoli and Curcio, 2006). Membrane distillation (MD), osmotic distillation (OD) and supported liquid membranes (SLMs) are typical examples of MCs. Their properties are summarized in Table 22.2.

Table 22.2 Basic properties of membrane contactors

Process	Driving force	Permeate	Retentate
MD	Temperature difference	Volatiles (water vapour)	Nonvolatiles
OD	Concentration difference	Volatiles (water vapour)	Nonvolatiles
SLMs	Concentration difference	Ions, low molecular weight organics, gases	Ions or less permeable organics/gases

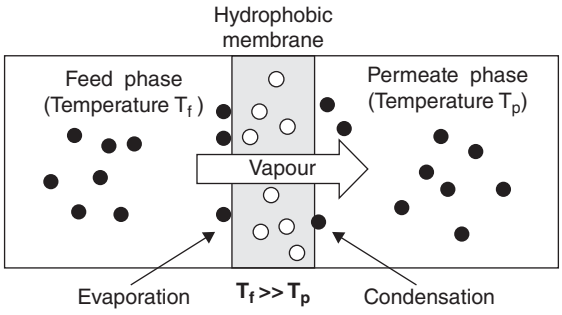

Figure 22.4 Schematic representation of the membrane distillation process

MD is a relatively new membrane process in which two aqueous solutions, at different temperatures, are separated by a macroporous hydrophobic membrane. In these conditions a net pure water flux from the warm side to the cold side occurs. The process takes place at atmospheric pressure and at temperatures that may be much lower than the boiling point of the solutions. The driving force is the vapour pressure difference between the two solution-membrane interfaces due to the existing temperature gradient (Figure 22.4).

The water transport through a MD membrane can be summarized in three steps:

- evaporation at the feed side of the membrane;
- transport of the vapour phase through the membrane pores;
- condensation of the vapour at the permeate side of the membrane (El-Bourawi *et al.*, 2006).

The difference between MD and OD is that in MD the physical origin of the vapour pressure difference is a temperature gradient rather than a concentration gradient. In OD, also known as isothermal membrane distillation or osmotic evaporation, the membrane separates two aqueous solutions with different osmotic pressures (a diluted aqueous solution from a concentrated osmotic solution). Due to the difference of the water activity between the two solutions, water is transported through the membrane, involving its evaporation in one side and further condensation in the other side (Hogan *et al.*, 1998) (Figure 22.5). The most suitable materials for MD and OD membranes include polyvinyldifluoride (PVDF), polytetrafluoroethylene (PTFE) and polypropylene (PP). The size of macropores can range between 0.2 and 1.0 μm. The porosity of the membrane will range from 60 to 80% of the volume and the overall thickness from 80 to 250 μm, depending on the absence or presence of support.

In general, the thinner the membrane and the greater the porosity of the membrane, the greater is the flux rate. The membrane configurations used

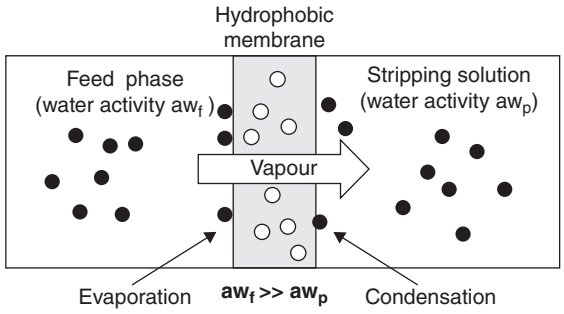

Figure 22.5 Schematic representation of the osmotic distillation process

include flat sheet, spiral wound and hollow fibre. Because MD and OD can be carried out at the atmospheric pressure and at a temperature which can be much lower than the boiling point of the solution, they can be used to concentrate thermosensitive solutes. Other advantages of MD and OD over other separation processes include less demanding mechanical property requirements and the possibility to reach high concentration levels. Therefore, MD and OD have received great attention as techniques for concentrating liquid foods, such as fruit juices (Jiao, Cassano and Drioli, 2004).

PVDF membranes showed very good retention towards orange juice compounds, such as total soluble solids, sugars and organic acids; the rejection of sugars and organic acids was 100%. On the other hand, a vitamin C degradation of 42.1%, mainly associated with a high temperature and oxidation, was observed. As a result, the operating temperature in the MD process must be maintained as low as possible (Drioli, Jiao and Calabrò, 1992). The OD process offers some potential advantages over RO and MD processes in the concentration of liquid foods. As already mentioned, RO suffers from high osmotic pressure limitations, while in MD some loss of volatile components and heat degradation may still occur due to the heat requirement for the feed stream in order to maintain the water vapour pressure gradient. OD does not suffer from any of these problems when operated at room temperature.

The typical OD process involves the use of a concentrated brine at the downstream side of the membrane as the stripping solution. $CaCl_2$ is commonly used because it is not toxic and is readily available at a low cost. Potassium salts of ortho- and pyrophosphoric acid are also indicated for their low equivalent weight, high water solubility, steep positive temperature coefficient of solubility and safe use in foods and pharmaceuticals (Hogan *et al.*, 1998).

The most well-known module designed for OD is the Liqui-Cel membrane contactor containing microporous PP hollow fibres. These fibres are approximately 0.3 mm in external diameter with a wall thickness of about 0.03 mm; they have a mean pore diameter of about 30 nm and a porosity of about 40%. The membrane module is characterized by a shell and tube configuration; in

most cases, the juice is recirculated in the shell side, while the brine solution is recirculated in the lumen side.

Several studies related to the OD concentration of single-strength and clarified fruit juices (orange, cactus pear, pineapple, passion fruit, melon, etc.) are reported in the literature (Galaverna *et al.*, 2008; Hongvaleerat *et al.*, 2008; Cassano and Drioli, 2007; Cassano *et al.*, 2007, 2003; Cissé *et al.*, 2005; Rodrigues *et al.*, 2004; Vaillant *et al.*, 2005, 2001). At a low total soluble solid concentration of the juice, the OD flux decay is more attributable to the reduction of the brine concentration, while it mainly depends on juice viscosity when juice concentration reached a value higher than 40 °Brix. Almost all physicochemical parameters of the juice were very well preserved in the concentrated juice. Furthermore, no significant loss of vitamin C and other antioxidant compounds was observed in comparison with juices concentrated by thermal evaporation.

Problems associated with commercial application of OD in concentrated fruit juice processing are the low evaporation fluxes compared with those of thermal evaporation (and hence higher production costs) and the management of the diluted brine strip. It is essential to reuse the brine several times before it is removed from the process. Corrosion and scaling make the regeneration process expensive for exhausted brines that can hardly be accomplished by conventional evaporators. In addition to heat evaporation, the effective methods need to be developed for the concentration of the osmotic brine. The key factors of conventional evaporation, RO and membrane contactors in the concentration of liquid foods are summarized in Table 22.3.

Other selected applications of membrane contactors in the food industry are the bubble-free carbonation of soft drinks, the adjustment of the alcohol content in wine and the CO_2 removal followed by nitrogenation in beer production. The idea to use a thin organic liquid layer separating aqueous phases is very attractive because the diffusion in liquids is much higher than in solid polymers and inorganic membranes (Liley, Reid and Buck, 1984). In

Table 22.3 Key factors of conventional evaporation and membrane concentration techniques (source: Adapted from Jiao *et al.* 2004. Reproduced with permission of Elsevier)

Process	Maximum achievable concentration (°Brix)	Product quality	Evaporation rate or flux	Operating cost	Capital investment	Energy consumption
Evaporation	60–70	Poor	200–300 L/h	Moderate	Moderate	Very high
Reverse osmosis	25–30	Very good	5 – 10 L/m^2 h	High	High	High
Membrane distillation	60–70	Good	1 – 10 L/m^2 h	High	Moderate	Low
Osmotic distillation	60–70	Very good	1 – 3 L/m^2 h	High	High	Low

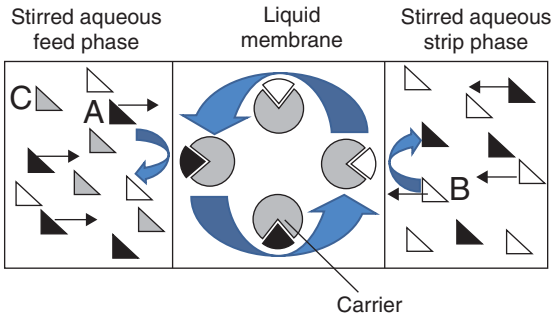

Figure 22.6 Schematic representation of supported liquid membranes

supported liquid membranes (SLMs), an organic liquid is imbedded in small pores of a microporous support used to separate two aqueous phases – the feed phase and the strip phase. The support contains a carrier able to bind the species of interest on the feed side and transfer it through the micropores on the strip side. Figure 22.6 shows the transport mechanisms of ions through a SLM. The charged species A is selectively extracted by the carrier through the organic membrane/feed interface. The carrier picks up A, moves across the membrane as a complex and exchanges A with the charged species B on the strip phase (Kocherginsky, Yang and Seelam, 2007).

Flat sheet membranes are typically used in small experimental laboratory plants consisting of two compartments separated by the membrane. Hollow fibre SLMs are also available; they offer a higher surface area per unit of module volume but their cost is relatively high. Usually SLMs are based on the use of an organic solvent containing a selective carrier immobilized in the micropores of a hydrophobic membrane separating two aqueous solutions. Hydrophilic membranes separating two non-aqueous phases impregnated by a water phase can also be used (Miyako *et al.*, 2003). The low stability of the membrane due to the volatility of water is the main problem in using this configuration. The use of ionic liquids seems to be a promising approach to overcome this drawback.

The extraction of valuable products from natural sources is an interesting perspective of SLMs for agro-food applications. For instance, the separation of organic acids obtained as by-products of fermentation processes from reagents and impurities is of great interest for biochemical and pharmaceutical applications. In this field, SLMs offer different advantages in terms of lower energy consumption, higher selectivity and faster separation rate over traditional methods, such as extractive fermentation and selective precipitation (Crespo and Böddeker, 1993). In fruit juice processing, interesting applications of SLMs are related to the extraction of organic acids from fruit juice (Schäfer and Hossain, 1996) and the reduction of bitter compounds in citrus juices.

22.6 Membrane bioreactors

Traditionally bioconversions are realized in classical batch reactors, which are limited by different factors such as high enzyme purification cost, low productivity per unit time, high labour cost and great variability of product quality and difficult and expensive recovery of enzymes or cellular microorganisms. The use of enzymes immobilized on solid supports leads to a reduction of the overall production cost due to the reutilization of the biocatalyst and the use of a continuous process.

Membrane bioreactors (MBRs) are systems in which bioconversions are integrated with selective membrane separations; they represent a valid alternative to classical methods of enzymatic immobilization (Figure 22.7). In these systems, the membrane assures the complete rejection of enzymes in the reaction vessel while the products are continuously extracted from the medium, reducing their inhibitory effect on the reaction rate. The most common configurations of MBRs can be classified into two types, as shown in Figure 22.7. In the first case, biocatalysts (e.g. enzymes, microorganisms and antibodies) are compartmentalized by the membrane in a reaction vessel so that the membrane does not contribute to the reaction but only controls the mass transport (Figure 22.7a). These systems are also called free enzyme membrane bioreactors (FEMBRs) or free cell membrane bioreactors (FCMBRs). Alternatively, enzymes can be immobilized on the membrane surface or within

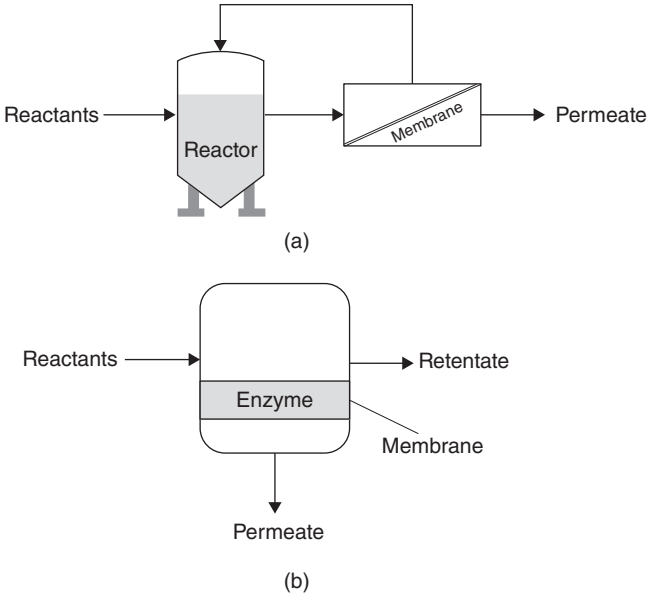

Figure 22.7 Schematic representation of (a) free enzyme membrane bioreactor (FEMBR) and (b) immobilized enzyme membrane bioreactor (IEMBR)

the membrane matrix itself (Figure 22.7b). In this case, the membrane acts as a support for the catalyst and as a separation unit (Giorno and Drioli, 2000). These systems are also called immobilized enzyme membrane bioreactors (IEMBRs).

The use of FEMBRs offers some advantages over the traditional batch reactor configuration in terms of continuous operating mode, high concentration of biocatalysts and continuous removal of inhibitors. As a consequence, high production rates can be achieved, ensuring the economic viability of the process. However, some drawbacks are related to the loss of bioactivity due to adsorption to the membrane surface as well as mechanical stress and membrane fouling, which reduces the membrane flux and the cost-efficiency of the process. The FEMBRs are particularly suggested when the substrate and enzyme are larger than the products. When the substrate and product show a similar size, the use of IEMBRs is preferred. Membranes used in MBRs can have a symmetric or an asymmetric structure. A wide range of polymeric membranes, including polysulfone (PS), polyethersulfone (PES), polypropylene (PP) and polyamide (PA) are used as well as inorganic membranes. They can be assembled in a plate-and-frame, spiral-wound or tubular configuration.

Enzymes and cells can be entrapped within the membrane structure during the preparation of the membrane by mixing the enzyme solution with the polymeric solution (Tan, Wang and Zhang, 2002). Alternatively, the enzyme can be immobilized by gelification on the membrane surface by filtering the enzyme solution within the porous layer of asymmetric hydrophilic membranes (Wang *et al.*, 2008). Finally, biocatalysts can be attached at the membrane surface through covalent or noncovalent (adsorption through hydrophobic or ionic interaction) bindings (Shamel *et al.*, 2007).

The use of MBRs in food applications concerns the processing of food and beverages and the production of food ingredients obtained by biocatalytic processes (e.g. sugars, organic acids, esters, etc.). The clarification of fruit juices by MF or UF is generally preceded by a pre-treatment with pectinases devoted to hydrolyse pectins which are responsible for membrane fouling. This operation is commonly realized in batch systems that do not permit the recovery of the enzyme; further, the use of immobilized enzymes is characterized by steric hindrance effects and losses in enzyme activity due to the immobilization.

The use of FEMBRs offers different advantages if compared with a batch system in terms of increasing enzyme utilization, enhancement of reactor productivity and a significant reduction of product inhibition. Bélafi-Bakó *et al.* (2007) reported that more than 40% higher productivity was achieved when pectin is hydrolysed by polygalacturonase from *Aspergillus niger* in an FEMBR equipped with a 30 kDa regenerated cellulose membrane in comparison with a batch system.

FEMBRs based on the use of pectinases are also used in wine clarification in order to improve processability (Rodriguez-Nogales *et al.*, 2008). Additionally, the use of glucosidases and pectinases for the production of aromatic

compounds, flavour and additives are other interesting potentialities of MBRs in wine processing. A well-established application of MBRs in the dairy industry concerns the lactose hydrolysis in milk and whey. This process is of interest in order to improve milk digestibility and to reduce lactose crystallization in the manufacture of refrigerated dairy products. Additionally, the lactose hydrolysis is associated with the production of food additives like lactic acid, glucose and galactose. An innovative process to produce lactose-reduced skim milk was proposed by Novalin, Neuhaus and Kulbe (2005). In this approach, skim milk was pumped in the lumen side of a hollow fibre membrane module while the enzymatic solution containing β-galactosidase was circulated in the shell side. Molecules with lower size than the membrane cut-off value, such as small proteins, salts and lactose, entered the shell side where lactose was enzymatically converted to glucose, galactose and a small amount of oligosaccharides. A conversion rate of 78.1% was achieved at a temperature of 23 ± 2 °C. Some examples of MBRs for the production of lactic acid by fermentation processes from cheese whey are also reported in the literature (Li and Shahbazi, 2006; Tango and Ghaly, 2002). Another interesting application of MBRs in the dairy industry is related to the hydrolysis of milk and whey proteins in active peptides recognized as potential modulators of many regulatory processes.

A process for the stable production of low allergenicity hydrolysates from whey proteins was studied by Guadix, Camacho and Guadix (2006) by using a bacterial protease in a continuous stirred tank membrane reactor including a PES plate and frame UF module with a molecular weight cut-off of 3 kDa. A hydrolysate with an average peptide chain length of around four amino acids was obtained. The antigenic whey protein in the product was reduced to 99.97%, which suggested that it can be incorporated as a nitrogen source in infant formula and enteral nutrition. Trusek-Holownia (2008) analysed the hydrolysis of casein in the presence of thermolysin and subtilisin in a MBR equipped with a flat sheet membrane. It was proved that at a relatively high substrate retention (i.e. ca. 50%), the substrate conversion degree could increase by about 50%. Owing to the high cost of biocatalysts such process intensification is significant.

MBRs play an important role also in sugar and starch processing. Starch hydrolysates are mainly produced through enzymatic degradation of starch masses and constitute a significant product utilized in the food industry (pure dextrose, high fructose syrups and sweeteners). At an industrial level, the enzymatic hydrolysis of starch is performed in batch reactors through a two-step procedure including liquefaction and saccharification. This approach is characterized by numerous disadvantages, such as incompatibility of enzyme recovery and reuse, high labour and purification costs, high capital investment and discrepancies in glucose syrup quality.

The coupling of enzymatic saccharification with a UF membrane in the same apparatus allows the separation of hydrolysis products with

simultaneous recycling of undigested substrate and enzyme, and removal of inhibitory products from the reaction media. FEMBRs using amylolytic enzymes or liquefied starch as substrate have been developed on a laboratory scale. For instance, Grześkowiak-Przywecka and Słomińska (2007) studied the hydrolysis of maltodextrin by a fungal α-amylase for the production of maltose syrup through the simultaneous use of a stirred tank reactor and ceramic UF membranes with different MWCO. The maximal maltose content in the permeate using a 5 kDa cut-off was 63%. The steady state of hydrolysis was achieved after a 2–3 h reaction.

The one-step hydrolysis of starch with termamyl enzyme in a continuous recycle membrane reactor was also studied by Paolucci-Jeanjean *et al.* (2000). In addition to the economy on enzyme and purification costs due to reaction and separation integration, the observed productivity was clearly higher than the one obtained in the batch process. The use of turbulence promoters or IEMBRs has been proposed (Sarbatly and England, 2004) in order to prevent membrane fouling, which is the major drawback in the application of the membrane reactors together with enzyme inactivation. The starch hydrolysis for the production of sugars (such as glucose and maltose) and cyclodextrins for use in the food industry (i.e. in the production of jellies and fruit desserts) by using MBRs has been also reported (Paolucci-Jeanjean *et al.*, 1999).

Finally, enzymatic MBRs can be successfully employed for the production of emulsifiers, aroma compounds, free fatty acids and mono- or diglycerides by using biocatalysts like lipases or esterases, which catalyse a wide range of reactions (hydrolysis, transesterification, enantiomer resolution, etc.). Some examples concern the hydrolysis of milkfat and sunflower oil in free fatty acids (Gan, Rahmat and Weatherley, 1998; Garcia *et al.*, 1992), the synthesis of esters such as butyl, butyrate and butyl laurate in organic solvents (Lozano *et al.*, 2004; Magnan *et al.*, 2004).

22.7 Pervaporation

Pervaporation (PV) is a membrane-based separation technique with promising applications in food technology. In this process, a liquid mixture is separated by partial vapourization through a dense perm-selective membrane resulting in a vapour permeate and a liquid retentate. The driving force of PV is a gradient in chemical potential generated by the application of a partial vapour pressure difference between the liquid feed and the vaporous permeate (Nagai, 2010; Karlsson and Trägårdh, 1996). In practice, this difference is created by reducing the permeate side vapour pressure by establishing a vacuum (Figure 22.8a). Alternatively, the partial vapour pressure difference can be generated by sweeping an inert gas on the permeate side (Figure 22.8b). In both cases, the vapour permeate is converted into a liquid by means of a condenser.

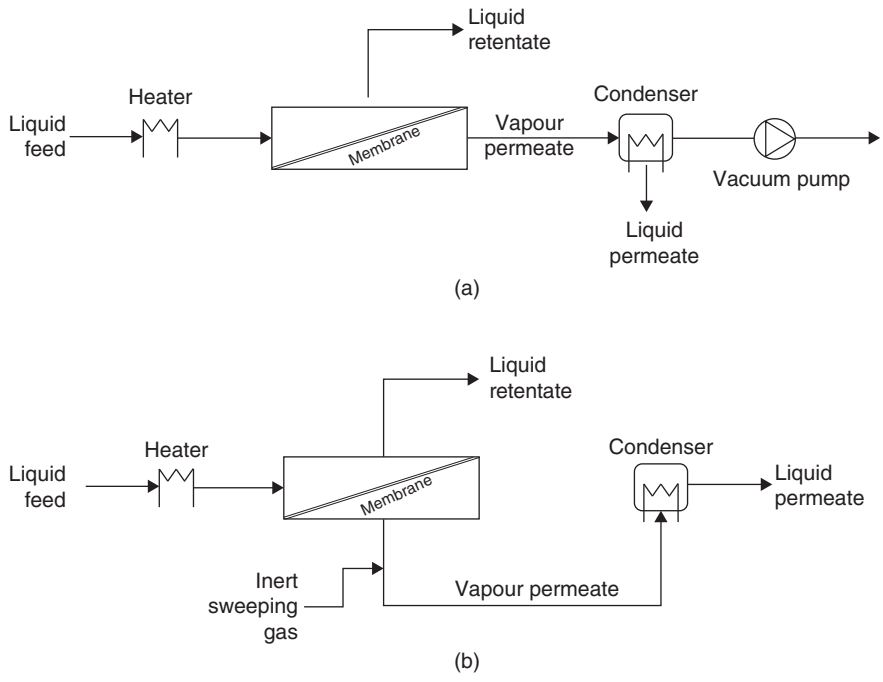

Figure 22.8 Schematic representation of (a) vacuum pervaporation and (b) sweep gas pervaporation

Polymeric membranes used in PV are usually manufactured as composites and can be either hydrophilic (such as those realized in poly(vinyl alcohol) and cellulose acetate) or hydrophobic (made in poly(dimethylsiloxane) or poly(trimethylsilypropyne)). Hydrophilic membranes permit preferentially the permeation of water while organic substances permeate preferentially through hydrophobic membranes. Ceramic PV membranes with a pore size of 0.5 nm prepared through an interaction of a pore structure (surfactant) and a silica sol have been also realized (Cuperus, 1998).

According to the solution-diffusion model, the mass transfer of the permeants across the membrane occurs in three steps: (1) a selective absorption of solutes from the bulk feed liquid into the membrane at the feed side, (2) a selective diffusion across the membrane owing the concentration gradient, and (3) a desorption of vapourous permeate solutes on the permeate side.

The mass flux across the membrane is inversely proportional to the membrane thickness. Compared with pressure-driven membrane operations, such as UF and RO, fluxes in PV are generally low, typically below 20 kg/m^2h. However, separation factors can be extremely high, often exceeding 1000. Applications of PV in food processing fall in one of the following areas: the recovery of aroma compounds from fruit juices, beer and plant extracts, wine or beer dealcoholization and processing of edible oils.

The recovery or concentration of aroma volatile compounds is a promising application of PV in the food industry. Quality degradation problems due to loss of aroma compounds and nutrients are typically encountered in the evaporative concentration processes of liquid foods. The use of PV hydrophobic membranes results in a permeate enriched in aroma compounds. By using vacuum PV, enrichment factors for aroma compounds greater than 100 have been obtained (Karlsson and Trägårdh, 1997) while operating at low temperatures (20 °C). This is a particular advantage of PV over other aroma recovery techniques operating at higher temperatures, especially when the aroma compounds of interest are susceptible to heat-induced deterioration.

Olefin PV membranes showed a good performance in the recovery of aroma compounds from tropical fruit juices due to their low water permeability and high enrichment factors to the components of aroma from both single strength and clarified fruit juices (Cardoso Pereira *et al.*, 2002). An integrated membrane process for the production of aroma-enriched apple juice concentrate was proposed by Álvarez *et al.* (2000). The process involved the use of an enzymatic membrane reactor to clarify the raw juice, a RO step to pre-concentrate the juice up to 25 °Brix, a PV step to recover and concentrate aroma compounds and a final evaporation step to concentrate the juice up to 72 °Brix. Polydimethylsiloxane (PDMS) and poly-octylmethylsiloxane (POMS) membranes in plate-and-frame configurations produced high overall enrichment factors in the range 100–1000 and overall mass transfer coefficients in the range 5–500 kg/m^2. The process performance was considerably influenced by operating parameters such as feed temperature, feed flow velocity and permeate pressure. Total flux before, during and after the experiments with apple juice did not change significantly, indicating low fouling problems for the investigated membranes in the selected operating conditions (20 °C, 11 000 Reynolds number and 2 mbar).

The dealcoholization of alcoholic beverages was one of the first applications of PV membranes in food processing (Brüschke, 1990). It is based on the use of hydrophobic membranes where ethanol permeates more readily than water. Since aroma compounds are much more hydrophobic than ethanol, they permeate together with ethanol; consequently, the flavour of the dealcoholized product is significantly reduced in comparison with the initial material. The use of hydrophilic membranes with low permeability towards hydrophobic compounds, such as aroma compounds, allows to overcome this drawback (Lee, Kalyani and Matson, 1991). Chardonnay wine was dealcoholized by using this process up to a final concentration of 0.5% (v/v), retaining most of the aroma compounds (> 80%).

An interesting application of PV in the processing of edible oils was proposed by Koseoglu *et al.* (1995). In this approach, a highly concentrated isopropanol–water mixture was used for oil extraction; then the oil–isopropanol–water mixture was evaporated, producing an oil phase (poor in isopropanol and water) and an isopropanol–water mixture, which

Table 22.4 Application of membrane technology in the food industry

Membrane process	Food area	Objectives
MF	Sugar	Removal of colour and particles
	Dairy	Clarification of cheese whey
		Defatting of milk
		Removal of bacteria from milk
		Milk standardization and concentration
	Fruit juice	Clarification of fruit juices
	Wine	Clarification of must and wine
	Beer	Clarification of beer
UF	Dairy	Fractionation of milk for cheese manufacture
		Fractionation of whey for whey protein concentrates
		Milk standardization and concentration
	Fruit juice	Clarification of fruit juice
	Wine	Clarification of must and wine
		Stability improvement of finished wine
	Beer	Clarification of beer
	Oil	Solvent recovery and particle removal
	Meat, fish and poultry	Protein recovery
	Sugar	Recovery of starch
NF	Dairy	Partial demineralization and concentration of whey
		Recovery of milk proteins and lactose
		Purification of oligosaccharides
	Fish	Brine cleaning
	Fruit juice	Recovery of phenols from fruit juices and by-products of fruit juice processing
RO	Fruit juice	Concentration of fruit juice
		Recovery of flavours, fragrances and pectins
		Recovery of sugars and acids from rinse water of fruit processing
		Recovery of caustic substance for peeling operations
		Recovery of limonine from orange juice
		Recovery of peach by-products
	Sugar	Concentration of maple syrup
	Fish	Concentration of water from processing of fish
	Vegetables	Separation of sugars from proteins in tomato serum
		Recovery of sweet potato stillage
		Recovery of soy and whey proteins
		Downstream process in corn refining
	Dairy	Concentration of milk or whey
		Polishing of NF permeates
	Wine	Removal of alcohol from wine
		Concentration of must
	Beer	Removal of alcohol from beer

Table 22.4 (*continued*)

Membrane process	Food area	Objectives
ED	Dairy	Demineralization and acidification of cheese whey
		Skimmed milk demineralization
	Sugar	Demineralization of molasses
		Demineralization of polysaccharides
	Fermentation	Demineralization of soy sauce
		Desalination of amino acids
		Recovering of organic acids
	Wine	Demineralization of must and wine
		Removal of tartrate from wine
	Fruit juice	Deacidification of fruit juice
MC	Fruit juice	Concentration of fruit juice
		Extraction of organic acids from fruit juice
		Recovery of aroma compounds
	Beer	Alcohol removal from beer
		CO_2 removal followed by nitrogenatation
		Deoxygenated water for the dilution of high-gravity beer
	Wine	Alcohol removal from wine
		Concentration of grape must
	Beverage	Carbonation of soft drinks
		Simultaneous oxygen removal
	Fermentation	Separation of organic acids from reagents and impurities
	Dairy	Concentration of whey proteins
MBR	Sugar	Production of glucose, fructose or maltose syrup from starch hydrolyses
		Production of maltodextrin or cyclodextrin from starch hydrolysates
		Production of pectic oligosaccharide or galacturonic acid from pectin hydrolysates
		Production of fructose syrup from sucrose hydrolysates
	Dairy	Production of peptides with low allergenicity or functional properties
	Oils and esters	Production of esters, fatty acids, monoglycerides and diglycerides
	Fermentation	Production of lactic acid, mannitol, xylitol and succinic acid
PV	Fruit juice	Recovery of aroma compounds
	Wine	Removal of alcohol
	Vegetables	Recovery of aroma compounds

was dewatered by PV and fed back to the extractor. This technique can be considered a valid alternative to the traditional extraction method based on the use of organic solvents (such as hexane and petroleum ether), which present serious drawbacks in terms of safety during the extraction processes and contamination of the final processed oil due to their toxicity. A summary of membrane applications in the food industry is shown in Table 22.4.

22.8 Conclusions

A general overview of potential and well-established applications of membrane technology in food processing is presented. Compared to traditional separation technologies, membrane processes show great flexibility, compactness and possibility of automation. Food safety, competitiveness and environmental friendliness are additional advantages.

Pressure-driven membrane operations today represent well-established molecular separation units in a wide range of applications in the food industries. A continuous growth of their market in food applications related to product streams, wastewater treatment and pre- and post-treatment of water is expected in the near future. Relatively new membrane operations, such as pervaporation, electrodialysis, membrane contactors and membrane bioreactors offer new possibilities in the production of food additives, recovery of high added-value compounds and aroma from food sources, and concentration of liquid foods. Finally, further technological inputs and economic benefits can be achieved through the integration of different unit membrane operations, between themselves or with other conventional technologies. In such a situation, the development of hybrid processes can give a significant contribution to redesign traditional flow sheets in the processing of liquid foods with consequent advantages in terms of improvement of food quality, recovery of by-products and high added-value compounds, control of environmental impact, reduction of energy and water consumption.

Abbreviations

ED	Electrodialysis
FCMBR	Free cells membrane bioreactor
FEMBR	Free enzymes membrane bioreactor
IEMBR	Immobilized enzyme membrane bioreactor
MBR	Membrane bioreactor
MC	Membrane contactor
MF	Microfiltration
MD	Membrane distillation
MWCO	Molecular weight cut-off
NF	Nanofiltration
OD	Osmotic distillation
PA	Polyamide
PDMS	Polydimethylsiloxane
PE	Polyethylene
PES	Polyethersulfone
POMS	Polyoctylmethylsiloxane
PP	Polypropylene
PS	Polysulfone

PTFE	Polytetrafluoroethylene
PV	Pervaporation
PVDF	Polyvinylidene difluoride
RO	Reverse osmosis
SLM	Supported liquid membrane
UF	Ultrafiltration
WPC	Whey protein concentrate

References

Al-Akoum, O., Ding, L.H. and Jaffrin, M.Y. (2002) Microfiltration and ultrafiltration of UHT skim milk with a vibrating membrane module. *Separation and Purification Technology*, **28**, 219–234.

Al-Akoum, O., Jaffrin, M.Y. and Ding, L.H. (2005) Concentration of total milk proteins by high shear ultrafiltration in a vibrating membrane module. *Journal of Membrane Science*, **247**, 211–220.

Álvarez, S., Riera, F.A., Álvarez, R., Coca, J., Cuperus, F. P., Bouwer, S., Boswinkel, G., van Gemert, R.W., Veldsink, J.W., Giorno, L., Donato, L., Todisco, S., Drioli, E., Olsson, J., Trägårdh, G., Gaeta, S. N. and Panyor, L. (2000) A new integrated membrane process for producing clarified apple juice and apple juice aroma concentrate. *Journal of Food Engineering*, **46**, 109–125.

Atra, R., Vatai, G., Bekassy-Molnar, E. and Balint, A. (2005) Investigation of ultra and nanofiltration for utilization of whey protein and lactose. *Journal of Food Engineering*, **67**, 325–332.

Bazinet, L. (2004) Electrodialytic phenomena and their applications in the dairy industry: a review. *Critical Review in Food Science and Nutrition*, **44**, 525–544.

Bélafi-Bakó, K., Eszterle, M., Kiss, K., Nemestóthi, N. and Gubicza, L. (2007) Hydrolysis of pectin by *Aspergillus niger* polygalacturonase in a membrane reactor. *Journal of Food Engineering*, **78**, 438–442.

Brüschke, H.E.A. (1990) Removal of ethanol from aqueous stream by pervaporation. *Desalination*, **77**, 323–329.

Cardoso de Oliveira, R., Caleffi Docê, R. and Davanted de Barros, S.T. (2012) Clarification of passion fruit juice by microfiltration: analyses of operating parameters, study of membrane fouling and juice quality. *Journal of Food Engineering*, **111**, 432–439.

Cardoso Pereira, C., Meira Rufino, J., Habert, A.C., Nobrega, R., Corrêia Cabral, L.M. and Borges, C.P. (2002) Membrane for processing tropical fruit juice. *Desalination*, **148**, 57–60.

Cassano, A. and Drioli, E. (2007) Concentration of clarified kiwifruit juice by osmotic distillation. *Journal of Food Engineering*, **79**, 1397–1404.

Cassano, A., Conidi, C. and Drioli E. (2010) Physico-chemical parameters of cactus pear (*Opuntia ficus-indica*) juice clarified by microfiltration and ultrafiltration processes. *Desalination,* **250**, 1101–1104.

Cassano, A., Donato, L. and Drioli, E. (2007) Ultrafiltration of kiwifruit juice: Operating parameters, juice quality and membrane fouling. *Journal of Food Engineering*, **79**, 613–621.

Cassano, A., Marchio, M. and Drioli, E. (2007) Clarification of blood orange juice by ultrafiltration: analyses of operating parameters, membrane fouling and juice quality. *Desalination*, **212**, 15–27.

Cassano, A., Drioli, E., Galaverna, G., Marchelli, R., Di Silvestro, G. and Cagnasso, P. (2003) Clarification and concentration of citrus and carrot juices by integrated membrane processes. *Journal of Food Engineering*, **57**, 153–163.

Cassano, A., Conidi, C., Timpone, R., D'Avella, M. and Drioli, E. (2007) A membrane-based process for the clarification and the concentration of the cactus pear juice. *Journal of Food Engineering*, **80**, 914–921.

Cheang, B. and Zidney, A.L. (2003) Separation of α-lactalbumin and β-lactalbumin using membrane ultrafiltration. *Biotechnology and Bioengineering*, **83**, 201–209.

Cheryan, M. (1998) *Ultrafiltration and Microfiltration Handbook*. Technomic Publishing Company, Lancaster.

Cissé, M., Vaillant, F., Perez, A., Dornier, M. and Reynes, M. (2005) The quality of orange juice processed by coupling crossflow microfiltration and osmotic evaporation. *International Journal of Food Science and Technology*, **40**, 105–116.

Crespo, J.G. and Böddeker, K.W. (1993) *Membrane Processes in Separation and Purification*. NATO ASI Series, Kluwer Academic Publishers, Dordrecht, The Netherlands.

Cuperus, F.P. (1998) Membrane processes in agro-food. State-of-the-art and new opportunities. *Separation and Purification Technology*, **14**, 233–239.

Daufin, G., Escudier, J.P., Carrère, H., Bérot, S., Fillaudeau, L. and Decloux, M. (2001) Recent and emerging applications of membrane processes in the food and dairy industry. *Transactions of the Institution of Chemical Engineers*, **79**, 89–102.

de Bruijn, J.P.F., Venegas, A., Martinez, J.A. and Bórquez, R. (2003) Ultrafiltration performance of Carbosep membranes for the clarification of apple juice. *LWT-Food Science and Technology*, **36**, 397–406.

Drioli, E., Criscuoli, A. and Curcio, E. (2006) *Membrane Contactors: Fundamentals, Applications and Potentialities*, Elsevier, Amsterdam, The Netherlands.

Drioli, E., Curcio, E. and Di Profio, G. (2005) State of the art and recent progresses in membrane contactors. *Chemical Engineering Research and Design*, **83**, 223–233.

Drioli, E., Jiao, B. and Calabrò, V. (1992) The preliminary study on the concentration of orange juice by membrane distillation. *Proceedings of the VII International Citrus Congress*, Acireale, Italy, vol. 3, pp. 1140–1144.

Drioli, E. and Romano, M. (2001) Progress and new perspectives on integrated membrane operations for sustainable industrial growth. *Industrial Engineering Chemical Resource*, **40**, 1277–1300.

El-Bourawi, M.S., Ding, Z., Ma, R. and Khayet, M. (2006) A framework for better understanding membrane distillation separation process. *Journal of Membrane Science*, **285**, 4–29.

Ernstrom, C.A., Sutherland, B.J. and Jameson, G.W. (1980) Cheese base for processing a high yield product from whole milk by ultafiltration. *Journal of Dairy Science*, **63**, 228–234.

Fillaudeau, L., Blanpain-Avet, P. and Daufin, G. (2006) Water, wastewater and waste management in brewing industries. *Journal of Cleaner Production*, **14**, 463–471.

Fukumoto, L.R., Delaquis, P. and Girard B. (1998) Microfiltration and ultrafiltration ceramic membranes for apple juice clarification. *Journal of Food Science*, **63**, 845–850.

Gabelman, A. and Hwang, S. (1999) Hollow fiber membrane contactors. *Journal of Membrane Science*, **159**, 61–106.

Galaverna, G., Di Silvestro, G., Cassano, A., Sforza, S., Dossena, A., Drioli, E. and Marchelli, R. (2008) A new integrated membrane process for the production of concentrated blood orange juice: effect on bioactive compounds and antioxidant activity. *Food Chemistry*, **106**, 1021–1030.

Gan, Q., Rahmat, H. and Weatherley, L.R. (1998) Simultaneous reaction and separation in enzymatic hydrolysis of high oleate sunflower oil-evaluation of ultrafiltration performance and process synergy. *Chemical Engineering Journal*, **71**, 87–96.

Garcia, H.S., Malcata, F.X., Hill, C.G., Amundson, C.H. (1992) Use of *Candida rugosa* lipase immobilized in a spiral wound membrane reactor for the hydrolysis of milkfat. *Enzyme and Microbial Technology*, **14**, 535–545.

Giorno, L. and Drioli, E. (2000) Biocatalytic membrane reactors: applications and perspectives. *Trends in Biotechnology*, **18**, 339–348.

Girard B. and Fukumoto L. (2000) Membrane processing of fruit juices and beverages: a review. *Critical Reviews in Food Science and Nutrition*, **40**, 91–157.

Grześkowiak-Przywecka, A. and Słomińska, L. (2007) Saccharification of potato starch in an ultrafiltration reactor. *Journal of Food Engineering*, **79**, 539–545.

Guadix, A., Camacho, F. and Guadix, E.M. (2006) Production of whey protein hydrolysates with reduced allergenicity in a stable membrane reactor. *Journal of Food Engineering*, **72**, 398–405.

Hogan, P.A., Canning, R.P., Peterson, P.A., Johnson, R.A. and Michaels, A.S. (1998) A new option: osmotic distillation. *Chemical Engineering Progress*, **94**, 49–61.

Hongvaleerat, C., Cabral, L.M.C., Dornier, M., Reynes, M. and Nigsanond, S. (2008) Concentration of pineapple juice by osmotic evaporation. *Journal of Food Engineering*, **88**, 548–552.

Jaffrin, M.Y., Ding, L.H., Al-Akoum, O. and Brou, A. (2004) A hydrodynamic comparison between rotating disk and vibratory dynamic filtration systems. *Journal of Membrane Science*, **242**, 155–167.

Jiao, B., Cassano, A. and Drioli, E. (2004) Recent advances on membrane processes for the concentration of fruit juices: a review. *Journal of Food Engineering*, **63**, 303–324.

Karlsson, H.O.E. and Trägårdh, G. (1996) Applications of pervaporation in food processing. *Trends in Food Science and Technology*, **7**, 78–83.

Karlsson, H.O.E. and Trägårdh, G. (1997) Aroma recovery during beverage processing. *Journal of Food Engineering*, **34**, 159–178.

Kelly, J. and Kelly, P.M. (1995) Nanofiltration of whey: quality, environmental and economic aspects. *Journal of the Society of Dairy Technology*, **48**, 20–25.

Khulbe, K.C., Feng, C.Y. and Matsuura, T. (2008) *Synthetic Polymeric Membranes, Characterization by Atomic Force Microscopy*. Springer Verlag, Berlin, Heidelberg, Germany.

Kocherginsky, N.M., Yang, Q. and Seelam, L. (2007) Recent advances in supported liquid membrane technology. *Separation and Purification Technology*, **53**, 171–177.

Koros, W.J., Ma, Y.H. and Shimidzu T. (1996) Terminology for membranes and membrane processes. *Journal of Membrane Science*, **120**, 149–159.

Koseoglu, S.S., Hernandez, E., Shah, V. and Tuohey, D. (1995) Opportunities for pervaporation: processing edible oils and fats. In: Bakish R. (ed.), *Proceedings of the Seventh International Conference on Pervaportaion Processes in Chemical Industry*. Bakish Material Corporation, Englewood, pp. 2–15.

Laorko, A., Zhenyu, L., Tongchitpakdee, S., Chantachum, S. and Youravong, W. (2010) Effect of membrane property and operating conditions on phytochemical properties and permeate flux during clarification of pineapple juice. *Journal of Food Engineering*, **100**, 514–521.

Lee, E.K., Kalyani, V.J. and Matson, S.L. (1991) Process for treating alcoholic beverages by vapour-arbitrated pervaporation. US Patent 5013447.

Li, Y. and Shahbazi, A. (2006) Lactic acid recovery from cheese whey fermentation broth using combined ultrafiltration and nanofiltration membranes. *Applied Biochemistry and Biotechnology*, **132**, 985–996.

Liley, P.E., Reid, R.C. and Buck, E. (1984) Physical and chemical data. In: Perry, R.H. and Green, D. (eds), *Perry's Chemical Engineers' Handbook*. McGraw-Hill Book Co., Singapore, p. 98.

Lipinzki, F. (2010) Cross-flow membrane applications in the food industry. In: Peinemann, K.V., Nunes, S.P. and Giorno, L. (eds), *Membrane Technology,* vol. **3**, *Membranes for Food Applications*, Wiley-VCH Verlag GmbH & Co., Weinheim, Germany, pp. 1–24.

Lonergan, D.A., Fennema, O. and Amundson, C.H. (1982) Use of electrodialysis to improve the protein stability of frozen skim milks and milk concentrates. *Journal of Food Science*, **47**, 1429–1434.

Lozano, P., Víllora, G., Gómez, D., Gayo, A.B., Sánchez-Conesa, J.A., Rubio, M. and Iborra, J.L. (2004) Membrane reactor with immobilized *Candida antarctica* lipase B for ester synthesis in supercritical carbon dioxide. *Journal of Supercritical Fluids*, **28**, 121–128.

Magnan, E., Catarino, I., Paolucci-Jeanjean, D., Preziosi-Belloy, L. and Belleville, M.P. (2004) Immobilization of lipase on a ceramic membrane: activity and stability. *Journal of Membrane Science*, **241**, 161–166.

Mallevialle, J., Odendaal, P.E. and Wiesner, M.R. (1988) *Water Treatment Membrane Processes*. McGraw-Hill Publishers, New York, USA.

Matta, V.M., Moretti, R.H. and Cabral, L.M.C. (2004) Microfiltration and reverse osmosis for clarification and concentration of acerola juice. *Journal of Food Engineering*, **61**, 477–482.

Merson, R.L., Paredes, G. and Hosaka, D.B. (1980) Concentrating fruit juices by reverse osmosis. In: *Ultrafiltration Membranes and Applications*. Plenum Press, New York, USA, pp. 405-413.

Miyako, E., Maruyama, T., Kamiya, N. and Goto, M. (2003) Use of ionic liquids in a lipase-facilitated supported liquid membrane. *Biotechnology Letters*, **25**, 805–808.

Mulder, M. (1991) *Basis Principles of Membrane Technology*. Kluwer Academic Publishers, Dordrecht, The Netherlands.

Nagai, K. (2010) Fundamentals and perspectives for pervaporation. In: Drioli E. and Giorno L. (eds), *Comprehensive Membrane Science and Engineering*, vol. **2**, *Membrane Applications in Molecular Separations*. Elsevier, Kidlington, Oxford, England, pp. 243–271.

Novalin, S., Neuhaus, W. and Kulbe, K.D. (2005) A new innovative process to produce lactose-reduced skim milk. *Journal of Biotechnology*, **119**, 212–218.

Paolucci-Jeanjean, D., Belleville, M.P., Rios, G. M. and Zakhia, N. (1999) Why on earth can people need continuous recycle membrane reactors for starch hydrolysis? *Starch/Stärke*, **51**, 25–32.

Paolucci-Jeanjean, D., Belleville, M.P., Rios, G.M. and Zakhia, N. (2000) The effect of enzyme concentration and space time on the performance of a continuous recycle membrane reactor for one-step starch hydrolysis. *Biochemical Engineering Journal*, **5**, 17–22.

Pepper, D. (1990) RO for improved products in the food and chemical industries and water treatment. *Desalination*, **77**, 55–71.

Rai, P., Majumdar, G.C., Sharma, S., Das Gupta, S. and De, S. (2006) Effect of various cutoff membrane on permeate flux and quality during filtration of mosambi (*Citrus sinensis* (L.) Osbeck) juice. *Food and Bioproduct Processing*, **84**, 213–219.

Rayess, Y.E., Bacchin, P., Taillandier, P., Raynal, J., Mietton-Peuchot, M. and Devatine, A. (2011) Cross-flow microfiltration applied to oenology: a review. *Journal of Membrane Science*, **382**, 1–19.

Rodrigues, R.B., Menezes, H.C., Cabral, L.M.C., Dornier, M., Rios, G.M. and Reynes, M. (2004) Evaluation of reverse osmosis and osmotic evaporation to concentrate camu-camu juice (*Myrciaria dubia*). *Journal of Food Engineering*, **63**, 97–102.

Rodriguez-Nogales, J.M., Ortega, N., Perez-Mateos, M. and Busto, M. (2008) Pectin hydrolysis in a free enzyme membrane reactor: an approach to the wine and juice clarification. *Food Chemistry*, **107**, 112–119.

Sarbatly, R. and England, R. (2004) Critical review of membrane bioreactor system used for continuous production of hydrolyzed starch. *Chemical and Biochemical Engineering Quarterly*, **18**, 155–165.

Schäfer, A. and Hossain, Md.M. (1996) Extraction of organic acids from kiwifruit juice using a supported liquid membrane process. *Bioprocess Engineering*, **16**, 25–33.

Shamel, M.M., Ramachandran, K.B., Hasan, M. and Al-Zuhair, S. (2007) Hydrolysis of palm and olive oils by immobilised lipase using hollow fibre reactor. *Biochemical Engineering Journal*, **34**, 228–235.

Strathmann, H., Giorno, L. and Drioli E. (2006) *An Introduction to Membrane Sciences and Technology,* Consiglio Nazionale delle Ricerche, Rome, Italy.

Tan, T., Wang, F. and Zhang, H. (2002) Preparation of PVA/chitosan lipase membrane reactor and its application in synthesis of monoglyceride. *Journal of Molecular Catalysis B: Enzymatic*, **18**, 325–331.

Tango, M.S.A. and Ghaly, A.E. (2002) A continuous lactic acid production system using an immobilized packed bed of *Lactobacillus helveticus*. *Applied Microbiology and Biotechnology*, **58**, 712–720.

Trusek-Holownia, A. (2008) Production of protein hydrolysates in an enzymatic membrane reactor. *Biochemical Engineering Journal*, **39**, 221–229.

Vaillant, F., Jeanton, E., Dornier, M., O' Brien G., Reynes, M. and Decloux, M. (2001) Concentration of passion fruit juice on an industrial pilot scale using osmotic evaporation. *Journal of Food Engineering*, **47**, 195–202.

Vaillant, F., Cisse, M., Chaverri, M., Perez, A.M., Dornier, M., Viquez, F. and Dhuique-Mayer, C. (2005) Clarification and concentration of melon juice using membrane processes. *Innovative Food Science and Emerging Technologies*, **6**, 213–220.

Vera, E., Sandeaux, J., Persin, F., Pourcelly, G., Dornier, M. and Ruales, J. (2008) Deacidification of passion fruit juice by electrodialysis with bipolar membrane after different pretreatments. *Journal of Food Engineering*, **90**, 67–73.

Vera, E., Sandeaux, J., Persin, F., Pourcelly, G., Dornier, M. and Ruales, J. (2009) Modeling of clarified tropical fruit juice deacidification by electrodialysis. *Journal of Membrane Science*, **326**, 472–483.

Vernhet, A., Pellerin, P., Belleville, M.P., Planque, J. and Montounet, M. (1999) relative impact of major polysaccharides on the performance of an organic microfiltration membrane. *American Journal of Enology and Viticulture*, **50**, 51–56.

Vladisavljević, G.T., Vukosavljević, P. and Bukvić, B. (2003) Permeate flux and fouling resistance in ultrafiltration of depectinized apple juice using ceramic membranes. *Journal of Food Engineering*, **60**, 241–247.

Wang, Y., Xu, J., Luo, G. and Dai, Y. (2008) Immobilization of lipase by ultrafiltration and cross-linking onto the polysulfone membrane surface. *Bioresource Technology*, **99**, 2299–2303.

23

Nanoparticles and Nanotechnology in Food

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23.1 Introduction

Nanotechnology is concerned with the characterization, fabrication and/or manipulation of structures or devices or materials that have at least one dimension (or contain components with at least one dimension) that is approximately 1–100 nm in length. A nanoparticle may also be defined as a material or a particle having all three of its external dimensions in the nanoscale. The behaviour, physical and chemical properties of the material at the nanoscale differ significantly from that of its macroscale form (Duncan, 2011; Chaudhry, Watkins and Castle, 2010). According to the National Nanotechnology Initiative (2006), 'Nanotechnology is the understanding and control of matter at dimensions of roughly 1 to 100 nanometers, where unique phenomena enable novel applications. Encompassing nanoscale science, engineering and technology, nanotechnology involves imaging, measuring, modeling, and manipulating matter at this length scale.'

23.1.1 Nanotechnology and food

The nanotechnology developments using nanodevices and nanomaterials have opened up new applications in agriculture, pharmaceutical and food sectors. Smart delivery systems, biosensors and nanoarrays have been developed for the agriculture sector. The food sector is expected to benefit through

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the use of smart biosensors, packaging materials and nanonutraceuticals (Kumari and Yadav, 2013).

The recent advancements in nanosciences and nanotechnologies are offering several new opportunities for innovation to food and allied industries. Though there are many potential benefits of these developments, there also exist a number of challenges to be solved. Many industrial and consumer sectors have identified nanotechnology as a new industrial revolution in the near future and the food sector is an obvious target of these new developments. Food-related applications of nanotechnology can offer a wide range of benefits to industries and consumers. It is possible to reduce the use of different food additives like preservatives, salt, fat and surfactants. The other important area is the development of new or improved tastes, textures and mouth feel through nanoscale processing. Further, nanoformulations can enhance the uptake, absorption and bioavailability of nutrients/supplements in the body compared to ongoing conventional bulk equivalents. Nanotechnology-derived polymer composites may offer new lightweight but tougher food packaging materials that can keep food products safe during transportation and extend the shelf-life (Chaudhry and Castle, 2011).

The subject 'food nanotechnology' is also associated with cell biology, food chemistry, biochemistry and nutrition, biophysics, and food technology and engineering. The main broader areas of food applications are on packaging, food safety, bioavailability, delivery system of nutrients, detection of pathogens, etc. Hence, it is desirable to understand the basic concepts that are linked with other subjects before performing the experiments related to nanotechnology, which is obviously an interdisciplinary subject by its understanding and application.

23.1.2 Food nanotechnology in nature

A few materials or substances in the nanoscale are present in nature and perform their functions in their original size in the human body. Nanosized structures are present in naturally occurring food substances and are also derived from natural raw materials. The proteins that are globular in nature have their diameter in the range of 10 to a few hundred nanometres. Polysaccharides, especially ribbon-shaped polymers, are less than 1 nm in thickness. Milk is another natural food, where the casein micelles are present in the form of nanostructures. The homogenization process reduces the fat globules in the milk to 100 nm. Physical, chemical and enzymatic treatments of whey protein can provide a novel nanotube. Natural proteins such as lactalbumin, collagen, gelatin, casein, zein, glutenin and gliadin are either used individually or in combination, and are manipulated to obtain a nanoparticle, of the size of several hundred nanometres. Carbohydrates like starch, cellulose, dextrin and their derivatives are also capable of forming a structure in nanoscale (Padua

and Nonthanum, 2012). Apart from these, simple triglyceride lipids are also among the naturally occurring food components that are only a few nanometres long, mostly about 2 nm. Finally, these proteins, carbohydrates and lipids are assimilated in the gastrointestinal system of the human body, after being broken down into nanostructures (Chaudhry, Castle and Watkins, 2010).

23.1.3 Available literature

Several books discuss the use of nanotechnologies in food (Bagchi *et al.*, 2013; Huang, 2012; Frewer *et al.*, 2011; Chaudhry, Castle and Watkins, 2010), health aspects (Malsch and Emond, 2014), general safety aspects (Asmatulu, 2013) and methodology involved (Padua and Wang, 2012). Review articles are also available on the development of nanoparticles (Moraru *et al.*, 2009; Kriegel *et al.*, 2008; Sanguansri and Augustin, 2006; Baeumner, 2004; Moraru *et al.*, 2003), applications of nanotechnology in food systems (Kumari and Yadav, 2013; Kuan *et al.*, 2012; Rashidi and Khosravi-Darani, 2011), food packaging applications of nanotechnology (de Azeredo, 2013; Dhall, 2013; Kalia and Parshad, 2013; Llorens *et al.*, 2012; Chaudhry and Castle, 2011; Duncan, 2011; Falguera *et al.*, 2011; Imran *et al.*, 2010; Rhim and Ng, 2007; Sorrentino, Gorrasi and Vittoria, 2007; Lopez-Rubio, Gavara and Lagaron, 2006), food analysis (Burris and Stewart, 2012), health aspects (Bouwmeester *et al.*, 2009; Shibamoto *et al.*, 2008; Vaclavik and Christian, 2008), regulatory aspects (Cushen *et al.*, 2012; Chau, Wu and Yen, 2007), safety aspects (Cockburn *et al.*, 2012; Magnuson, Jonaitis and Card, 2011; FAO/WHO, 2010; Bouwmeester *et al.*, 2009), nanofibres and nanotubes (Kriegel *et al.*, 2008; Graveland-Bikker and de Kruif, 2006), functional foods for health promotion applications (Chen, Weiss and Shahidi, 2006; Weiss, Takhistov and McClements, 2006), nanoscale properties of food materials (Lee *et al.*, 2007), structural design principles for delivery of bioactive components (Huang, Yu and Ru, 2010; McClements *et al.*, 2009), toxic potential of nanomaterials (Nel *et al.*, 2006) and incorporation of functional ingredients into foods by encapsulation (Gutiérrez *et al.*, 2013; Fathi, Mozafari and Mohebbi, 2012; Huang, Given and Qian, 2009; Pegg and Shahidi, 2007).

The present chapter thus discusses the advantages of nanotechnology, major developments and the methods involved in the synthesis of nanoparticles, application of nanotechnology for food processing and preservation in the agri-food sector, and risks associated with health hazards and toxicity regulations.

23.2 Advantages of nanotechnology

The major application of nanotechnology in food and nutrition is to design and develop novel functional food ingredients with improved characteristics

like water solubility, thermal stability, bioavailability, sensory characteristics and physiological performance. The specific advantages of nanotechnology for which it is being critically acclaimed are:

(a) control of physical and chemical properties to achieve targeted function,
(b) high surface area and/or interfacial (contact) area to mass and/or volume ratio enhances the taste, flavour and other transport parameters,
(c) better dispersability of water-insoluble substances in the food matrix without using other food additives,
(d) control over the solubility, retention time, release for a specified duration and site-specific or targeted release of a substance present in the food matrix,
(e) enhanced absorption, biological activity, bioreactivity and bioavailability of the material,
(f) better control over the size and size distribution, surface charge, surface chemistry, shape, lattice structure and arrangement, and
(g) ability to withstand mechanical, thermal and other stresses.

The main advantages that nanotechnologies offer over other existing technologies arise from the improved or novel functionalities of nanosized materials and substances. The very small size of nanomaterials enables dispersion of water-insoluble additives (such as colours, flavours and preservatives) in food products without the need for additional fat or surfactants. Nanosizing of bioactive substances is also claimed to give higher uptake, absorption and bioavailability in the body compared with bulk equivalents (FAO/WHO, 2010). It is for these advantages that the nanotechnology and nanoparticles are gaining importance and are being applied in several aspects of food (Bagchi *et al.*, 2013; Huang, 2012; Chaudhry, Castle and Watkins, 2010).

23.3 Applications in food preservation and processing

Nanotechnology has found several applications in the food sector by utilizing the different properties of nanomaterials. It varies from the antibacterial nanocoatings on food preparation surfaces that can help to maintain hygiene during food processing to the use of 'Smart' labels that can help to protect the safety and authenticity of food products in the supply chain (Chaudhry and Castle, 2011). The use of synthesized nanomaterials and the influence of nanotechnology on naturally occurring substances have developed many novel applications of which the recent developments that have happened in the last five years are shown in the Table 23.1.

The main applications include food packaging, preservation and improved nutritional benefits. However, applications involving food analysis, safety, colour and enhancing the textural and rheological properties of food have

Table 23.1 Applications of nanotechnology in different areas of food preservation and processing, and analysis

Broad area of application	Product and/or process	Materials and/or application	Reference
Food packaging	Antibacterial film	Zinc oxide and nisin on poly lactic acid and glass	Jin and Gurtler (2011)
	Antimicrobial films	Silver–gelatin nanoparticles	Halder *et al.* (2011)
	Antibacterial nanocomposites	Silver nanoparticles in hydroxypropyl methylcellulose	Moura, Mattoso and Zucolotto (2012)
	Antimicrobial peptide and biodegradable films; nanoencapsulation	Nisin	Imran *et al.* (2012)
Health benefits and food supplement, and fortification	Nanoencapsulation	Xanthophyll	Wang, Chen and Shi (2013)
	Nanoencapsulation	Bovine lactoferrin	Balcão *et al.* (2013)
	Nanoencapsulation	Beta-carotene	Gutiérrez *et al.* (2013)
Food preservation; extended shelf-life and improved stability of flavour in food	Nanoencapsulation; antimicrobial activity	Essential oils (D-limonene)	Donsi *et al.* (2011)
	Nanowebs/nanofibres	Vanillin–cyclodextrin complex in polyvinyl alcohol matrix	Kayaci and Uyar (2012)

(*continued overleaf*)

Table 23.1 (*continued*)

Broad area of application	Product and/or process	Materials and/or application	Reference
Functional food	Nanoencapsulation; functional bread;	Omega-3 poly unsaturated fatty acids	Gökmen *et al.* (2011)
Food analysis	Nanotubes and solid phase microextraction	Carbon nanotubes for detection of pthalates in beverages	Li *et al.* (2013)
	Nanotubes and solid phase microextraction	Carbon nanotubes for analysing carbamate pesticides in apples	Song, Shi and Chen (2013)
	Nanoparticles	Silica nanoparticles for determination of lead contamination in foods	Aboufazeli *et al.* (2013)
Food safety	Nanofibrous composites; antibacterial activity	Nano-zinc oxide in chitosan and polyvinyl alcohol	Wang *et al.* (2012)
Food colour	Nanoparticles	Beta-carotene in bivalent calcium ion cross-lined alginic acid	Astete *et al.* (2009)
Food texture and rheology	Nanostructure	Butter with improved stability and plasticity	Ivanov and Rashevskaya (2011)

also attained momentum in the recent years. The major applications are discussed in the subsequent sections.

23.3.1 Food packaging

The new applications so far appear to be on food packaging and health promoting food products, with only a few known examples in the mainstream of food processing and preservation. According to market estimates, food packaging applications make up the largest share of the current and short-term predicted market for nano-enabled food products. The promising area of application is the 'active' and 'smart' packaging systems (Chaudhry and Castle, 2011).

Nanocomposite applications as barriers, coating, release device and novel packaging systems are possible by modifying the permeation behaviour of foils, increasing barrier properties (chemical and microbial), improving mechanical and heat-resistance properties, developing active antimicrobial surfaces, sensing as well as signalling microbiological and/or biochemical changes and developing dirt-repellent coatings for packages.

The important functional roles of food packaging are to protect food from the surrounding system, contain the food, provide nutritional information for consumers and extend food shelf-life. The use of nanoparticles can improve the mechanical and heat resistance properties of food packaging and therefore increases the shelf-life by affecting gas or water vapour permeability. For example, polymers are not inherently impermeable to gases or water vapour, but polymer silicate nanocomposites have improved gas barrier property in addition to improved mechanical strength and heat resistance properties. The first nanocomposite was inspired by interacting an organic substance (protein, peptide or lipid) with an inorganic one (e.g. calcium carbonate) to form a material with increased toughness. Another example is a packaging material composed of potato starch and calcium carbonate. This foam, which is thermally stable and biodegradable, can replace the polystyrene used for fast food (Rashidi and Khosravi-Darani, 2011; Moraru *et al.*, 2003).

Generally, nanotechnology offers three possible distinct advantages with regard to food packaging, such as (a) acting as a barrier or provide resistance and improvement of functional properties, (b) incorporating active components to provide functional performance and (c) sensing relevant information about food quality (Avella *et al.*, 2013).

23.3.2 Nutraceutical/nutrient/supplement delivery systems

Food is often supplemented with nutrients of various types. Phytochemicals are food supplements with health benefits and are frequently used as part of

a diet. However, many phytochemicals are poorly absorbed by the human body because of low solubility and thus one of the most important and interesting applications for encapsulation of phytochemicals is to enhance their bioavailability by changing the pharmacokinetics and biodistribution characteristics. Several researches have been carried out to study the health promotion properties of different phytochemicals, including devising novel encapsulation materials and methods with an overall intention of trying to enhance the nutritional quality as well as the stability of the functional ingredients.

Nanoemulsion-based delivery systems proved to be one of the pragmatic approaches to enhance oral bioavailability and biological efficacies of different phytochemicals (Huang, Yu and Ru, 2010). These can offer advantages to food industries as many food-grade lipids and emulsifiers are available, and the processes become simpler and easier compared to conventional other lipid-based delivery systems. However, for the majority of delivery systems, the *in vivo* biological efficacies of encapsulated phytochemicals remain mostly unknown. Hence, there is a scope for developing novel value-added food-grade or GRAS materials from biomass, as well as understanding the potential impacts of these nanoencapsulated nutraceuticals on the human body.

Nanoencapsulation of nutraceuticals Improvement occurs for the water dispersability of many crystalline phytochemicals, such as beta-carotene and curcumin; also an improved *in vitro* anticancer activity has been reported (Gutiérrez *et al.*, 2013; Ofori and Hsieh, 2013). During the past decade, much effort has been devoted to the design and development of different nutraceutical delivery systems, and significant progress has been achieved. A number of delivery systems with different structures are now available. Nanoencapsulation for controlled release of nutrients, proteins, antioxidants and flavours are possible (Kuan *et al.*, 2012). Fortification of food by omega-3 fatty acid, lycopene, beta-carotene phytosterols and DHA/EPA (docosahexaenoic acid/eicosapentaenoic acid) are a reality with nanotechnology.

Therefore, more effort needs to be devoted to the development of novel value-added food-grade or GRAS materials from biomass, as well as an understanding of the potential impacts of these nanoencapsulated nutraceuticals to human health.

23.3.3 Food sensing and safety

Detection of very small amounts of chemical contaminants and residues is possible with the application of nanotechnology. Specific examples are:

(a) monitoring and tagging of food ingredients,
(b) use of electronic nose and tongue for sensory evaluation and

(c) food-borne pathogen identification by measuring nucleic acid, protein or any other indicator metabolite produced by microorganisms (Rashidi and Khosravi-Darani, 2011).

23.3.4 Biopolymers

Among polymeric materials and composites, a rapid growth in the use of polymer nanocomposites (PNC) has been reported though their applications in food packaging are still at the developmental stage. Simple conventional packaging material can be replaced with multifunctional intelligent packaging to maintain good quality during storage. Novel packaging solutions in this area will increasingly focus on food safety by controlling microbial growth, delaying oxidation and improving tamper visibility. Conventional composites usually require a high content of the inorganic filler phase to induce noticeable improvements in mechanical, thermal and barrier properties. Nanocomposites can achieve similar or better properties, such as increased tensile strength and improved thermal stability, etc., with a low proportion of filler (1–5%). The developed materials possess a lower density and higher processability than those obtained through the addition of traditional reinforcements. Common examples include isotactic polypropylene (iPP) filled with $CaCO_3$ nanoparticles, starch filled with clay nanoparticles, and polyethylene terephthalate (PET) filled with $CaCO_3$ nanoparticles (Avella *et al.*, 2013).

Nanolaminates consist of two or more layers of nanomaterials (physically or chemically bonded to each other) and are suitable for use in the food industries. Nanolaminates can be used for the preparation of edible coatings and films as well as foaming, which are currently applied in food industries for fruit, vegetables, meat, chocolate, candies, bakery products and French fries. These coatings or films are suitable as barriers to moisture, lipids or gases and increase the textural properties of foods, or are applied as carriers of functional agents including colours, flavours, antioxidants, nutrients and antimicrobials. At present, proteins, polysaccharides and lipids are being used to produce these films and coatings. Nanolaminates are also used as the coating material on food surfaces due to their low thickness and fragile characteristics (Rashidi and Khosravi-Darani, 2011).

23.3.5 Nanosensors and nanobiosensors

Nanotechnology has played a key role in the development of new generation diagnostic devices and novel sensors that are used to improve the quality of food and tracking of food products through the supply chain. Nanobiosensors

have been used for detecting both chemical and bacterial contaminants (del Carlo and Compagnone, 2008).

Nanosensors allow monitoring of various components from the time the food enters the food supply chain until it is consumed. Sensors based on microfluidic devices have miniaturized sensing systems and improved sensitivity, and enable real-time detection of pathogens and contaminants (Baeumner, 2004). Fluorescent semiconductor quantum dots have been used for detection of food-borne pathogens. An example is the use of quantum dots for detection of *Salmonella typhimurium* in chicken carcass wash water (Yang and Li, 2005). Among other nanosensor developments are the array biosensors for detection of food-borne contaminants, devices with nanoelectromechanical system (NEMS) technology for detection of pathogens, carbon nanotube-based sensors for detection of capsaicinoids in chilli peppers, electronic tongues and noses to detect compounds arising from deterioration of foods and nanocantilevers for recognition of proteins and pathogen detection (Augustin and Oliver, 2012; Sozer and Kokini, 2009).

Incorporation of nanosensors into food packaging or food premises gives producers, distributors and consumers real-time nutrition status of foods and alarms them prior to food deterioration. In addition, nanosensors provide a rapid, simple, cheap and accurate determination of chemical and microbial contamination, as compared to the conventional methods such as high-performance liquid chromatography (Nachay, 2007). Nanosensors are also able to detect environmental changes such as temperature, humidity and gas composition, metabolites from microbial growth and by-products from food degradation.

23.3.6 Biofilms

The development of biofilms using different food ingredients is an interesting area of research. The use of the bio-based polymeric materials including proteins, polysaccharides, lipids and other polymers of biological origin in food packaging is increasing because they have many characteristics relevant for technological applications and they are amenable to biodegradation (Halder *et al.*, 2011). The role of nanotechnology is to form a nanobiocomposite on combining a nanoparticle with a natural polymer that can give filming characteristics (Falguera *et al.*, 2011).

Filters with nanopores can find applications in purification of water to remove pathogens and other contaminants, and for developing emulsions with uniform size (Bouwmeester *et al.*, 2009). The commercial use of edible films has been limited due to problems related to their poor mechanical and barrier properties when compared to synthetic polymers. Several nanocomposites have been developed by adding reinforcing compounds (nanofillers) to biopolymers, improving their properties and efficiency (Falguera *et al.*, 2011).

Conventionally, mineral fillers such as clay, silica and talc have been incorporated in film preparation in the range of 10–50% (w/w) in order to reduce its cost or to improve its performance (Rhim and Ng, 2007). Thus, the most important nanoparticles that have been used to provide enhanced properties to edible films are clays. The nanometre-size dispersion of polymer-clay nanocomposites exhibit significant improvement in the mechanical and physical properties compared with pure polymer or conventional composites. Both proteins and polysaccharides have given rise to films in combination with nanoclay particles. However, other nanoparticles such as tripolyphosphate-chitosan, microcrystalline cellulose and silicon dioxide have also been added to biopolymers to obtain films. These nanoparticles can improve moisture barrier properties and restrict microbial growth (Falguera *et al.*, 2011).

23.3.7 Nanoscale enzymatic reactor

Enzymes are biological catalysts and control most of the biochemical reactions occurring in living cells. Difficulties for their industrial scale utilization have been attributed to a loss of activity in environments different from those in which they normally function. Only a limited number of studies on enzyme electrospinning has been published, despite the great appeal that such structures can serve as mini reactors. In most cases, these bioactive compounds are attached to prefabricated nanofibres through adsorption and subsequent covalent linkage of the enzyme to the surface of the fibres. Examples of enzymes attached to surfaces include lipase and catalase. Sawicka, Gouma and Simon (2005) have described the successful electrospinning of polyvinyl pyrrolidone with urease. The enzyme activity has been retained even after processing in the high electrical field, producing nanofibres of 7–100 nm intersected by spherical urease aggregates of 10–800 nm. The large surface area of the nanofibres markedly improves the reaction rate. Composite fibres containing 1.3% chymotrypsin have an average fibre diameter of 815 ± 190 nm. Interestingly, the relative bioactivity of the enzyme is better retained in nanofibres over the course of two weeks when compared with the bioactivity of unencapsulated chymotrypsin, which rapidly decreases below its detection limit within one day (Padua and Nonthanum, 2012).

23.3.8 Enhanced heat and shock resistance of packages

Biodegradable packaging materials with enhanced barrier characteristics and strength have been developed via the combination of nanoscale inorganic material and natural biopolymers. The diverse side chains and functional groups of these biopolymers offer greater flexibility in designing new packaging material to suit the properties of finished products. Biopolymersilicate nanocomposites with enhanced barriers, strength and functionalities have

been developed via the introduction of inorganic silicates or clays into a biopolymeric matrix. The addition of 3–5% of clay into bionanocomposites is sufficient to produce a lighter, stronger and more heat-resistant packaging, in addition to serving as an improved oxygen, carbon dioxide, moisture and volatile barrier (Kuan *et al.*, 2012).

A 5% clay/thermoplastic starch nanocomposite developed by Park *et al.* (2003) has shown a higher tensile strength and lower water vapour transmission rate than the clay-free counterpart. Miyagawa *et al.* (2005) have synthesized novel bio-based nanocomposites from 20% functionalized vegetable oil and 5% organically modified layered silicate clay with a better elastic modulus as compared to other bio-based epoxy composites containing the same amount of functionalized vegetable oil. A high clarity, light, strong and heat-resistant silicate–polyamide nanocomposite has also been developed to be suitable for fresh meat and other foods (Kuan *et al.*, 2012).

23.3.9 Nanoencapsulates

Functional materials in foods are incorporated and protected in a delivery system starting from the time of synthesis, storage, ingestion and, lastly, absorption into our bodies. A good delivery system is required to maintain these functional materials in an active state and release them to the desired physiological sites at the desired time and rate. Nanometre-sized association colloids, lipid-based nanoencapsulators, nanoemulsions, biopolymeric nanoparticles, nanolaminates and nanofibers are the delivery systems that have been developed for the purpose of encapsulation, protection and targeted release of food materials (Kuan *et al.*, 2012).

Most of the association colloids are nanoparticles with sizes ranging from 5 to 100 nm (Weiss, Takhistov and McClements, 2006). Micelles and vesicles are spherical nanoparticles with a diameter of 50 to 100 nm (Sanguansri and Augustin, 2006). They are often applied as delivery systems for insoluble compounds via entrapment within the hydrophobic interiors of the structure (Weiss, Takhistov and McClements, 2006). Successful applications include the encapsulation of limonene, lycopene, lutein, phytosterols, omega-3 fatty acids and DHA/EPA into canola cooking oil. Liposomes, archaeosomes and nanocochleates are a few examples of lipid-based nanoencapsulators (Kuan *et al.*, 2012; Joseph and Morrison, 2006).

Biopolymeric nanoparticles are biodegradable and possess spontaneous self-assembly ability to form a stable compact structure (Weiss, Takhistov and McClements, 2006). Zein, casein, chitosan, polylactic acid, poly-3-hydroxybutyrate and polycaprolactone have been found suitable for nanoencapsulating functional ingredients and releasing them to physiological target sites (Kuan *et al.*, 2012; Sanguansri and Augustin, 2006).

Nanolaminates are thin films formed by multiple nanolayers of carbohydrates, proteins, charged lipids or colloidal particles coated on a charged

surface through mainly electrostatic attraction of opposite surface charges (Weiss, Takhistov and McClements, 2006). In addition to their function in food packaging as edible coatings that prevent moisture, lipids and gas diffusion in fruits, vegetables, meats, confectionery and bakery products, nanolaminates can also serve as the carrier for bioactive compounds such as antibrowning agents, enzymes, colours, flavours, antioxidants and nutrients (Kuan *et al.*, 2012).

A new plastic nanocomposite has been developed to replace the conventional plastic bottle of beer with improved shatter-proof properties. The embedded nanoclay prevents oxygen and gas diffusion into or from the interior of the bottle and eliminates the issues of oxidative spoilage as well as flavour loss (Joseph and Morrison, 2006).

23.3.10 Metal nanoparticles

Nanoparticles made from metals and metal oxides have become the focus of intense research activity because of their improved physical, chemical and biological properties compared to their bulk counterparts. Heavy metals are usually toxic and highly reactive with proteins, particularly at the sulfhydryl groups. These metals possibly bind with different protein molecules and thus inhibit the regular metabolic processes of microorganisms. Among these heavy metals, gold and silver have been known to have extraordinary inhibitory and bactericidal properties since ancient times; these are also relatively nontoxic to the human cell up to a certain limit. Because of these characteristics, silver in various forms is suited for a wide range of applications in consumer, industrial and medical products. It is expected that in the nanoscale form, such metals or metal oxides can exhibit higher antimicrobial efficacy because they have a greater surface area compared to their bulk counterpart (Halder *et al.*, 2011). These authors have reported nanoscale silver particles stabilized by gelatin. These have been prepared through the reduction of aqueous silver nitrate solution by sodium borohydride. Transmission electron micrographs also indicate the presence of nanoscale silver particles of approximately 3.9 nm. The nanocomposites have exhibited significant antibacterial and antifungal activity.

Nanosilver particles are generally smaller than 100 nm and contain 20–15000 silver atoms. Silver nanoparticles have been receiving considerable attention as a result of their unique physical, chemical and biological properties, and have found important applications in optics, electronics and medicine. In addition, nanosilver has innate antimicrobial and antiparasitic activity (Singh, Kulkarni and Dash, 2013). Ahmad *et al.* (2003) and Nanda and Saravanan (2009) have also synthesized silver nanoparticles. These silver nanoparticles have found significant applications in a wide spectrum of biomedical utilities such as imaging and therapeutics, and especially as antimicrobial agents. However, silver is a heavy metal and raises controversy

upon application in food products. A cheaper and safer substitute for silver nanocomposites has subsequently been developed, using zinc oxide and magnesium oxide nanoparticles (Kuan *et al.*, 2012).

Oxygen scavenging ability of food packaging is desired to extend the shelf-life of products affected by direct oxidation, rancidity and spoilage by aerobic food microorganisms. Titanium nanoparticles incorporated into food packaging materials are able to scavenge oxygen. Titanium reacts with oxygen and moisture to form activated oxygen, leading to reduced concentration of oxygen within packaged foods (Xiao-e *et al.*, 2004). Other metals used include nanosilica, nanoselenium, nanotitanium dioxide, nanocalcium, nanoiron, etc. (FAO/WHO, 2010).

23.3.11 Nanoemulsions

Nanoemulsions are fine, oil-in-water emulsions with a mean droplet size of 50–200 nm. They do not scatter visible light and are hence transparent; these nanoemulsions remain stable for long periods owing to their small particle size. The bioavailability of lipophilic substances may be increased considerably by means of nanoemulsions. They have been in use for some time in parenteral nutrition. They also show unique textural properties, even at a low oil concentration, and have the consistency of a viscous cream, which makes them interesting for the development of reduced fat products (Padua and Nonthanum, 2012).

Nanoemulsion comprises of a kinetically stabilized dispersion of two or more immiscible liquids with dispersed phase droplets diameter between 50 to 1000 nm (Sanguansri and Augustin, 2006). Gu, Decker and McClements (2005) have synthesized a nano-structured multilayer emulsion that is stabilized by beta-lactoglobulin, iota-carrageenan and gelatin in multilayer membranes of dispersed oil droplets. Successful applications of nanoemulsions include bottled drinking water and milk fortified with vitamins, minerals and antioxidants (Gu, Decker and McClements, 2005). These functional ingredients are incorporated without affecting the organoleptic properties of the products, yet with controlled release of bioactive compounds (Sanguansri and Augustin, 2006).

23.3.12 Nanofibres

Nanofibres possess unique characteristics that result in their various functionalities. A high surface area to volume ratio provides a large reaction platform while the longitudinal rod shape allows pack alignment for

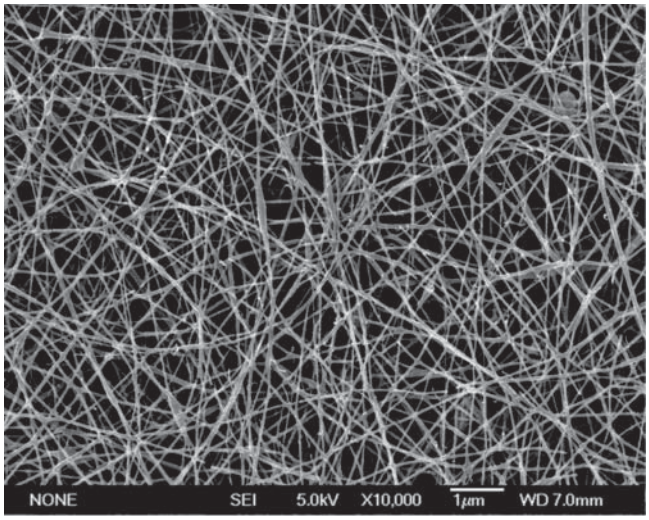

Figure 23.1 SEM image of chitosan-PEO composite nanofibres with a magnification of ×10 000 (source: Kriegel *et al.* 2008. Reproduced with permission of Taylor & Francis Ltd.)

superior mechanical properties such as stiffness and tensile strength. These characteristics make nanofibres suitable for use in targeted delivery systems, tissue engineering, wound dressing, composite reinforcement materials and carriers for catalysts and enzymes.

The nanofibres (Figure 23.1) have potential uses in the food packaging area, because these chitosan-based films are biodegradable and antimicrobial in nature. The ingredients used for developing nanofibres are mainly chitosan and polyethylene oxide (PEO). It can be used as a food preservative due to its broad range of antimicrobial activity against various microorganisms, such as fungi, yeast and bacteria. The nanofibres (Figure 23.1) are produced by a technique known as electrospinning (Kreigel *et al.*, 2008).

Electrospinning has recently generated much attention as a relatively cost-effective method to produce long and continuous nanopolymer strands. Nanofibres are produced through an electrically charged jet of a polymer solution or polymer melt. The polymer solution is spun from a small capillary orifice when the potential difference between the pipette tips and grounded collector surface is higher than the viscosity and surface tension of the polymer dispersion in the pipette. The polymer jet is elongated and stretched into a finer fibre before reaching the grounded collector plane, forming a nonwoven nanofibre mat (Kuan *et al.*, 2012).

23.3.13 Other applications

The other applications of nanotechnology in the field of food science and technology are:

- nanoceramic pan to reduce the time of roasting and amount of consumed oil, reduction of trans fatty acids due to usage of plant oil instead of hydrogenated oil, finally resulting in safe nano food development of nanocapsules that can be incorporated into food to deliver nutrients,
- nanomicelles for targeted delivery of nutrients (nutrition nanotherapy),
- selective passage of materials on the basis of shape and size (nanofiltration),
- enzyme and protein evaluation as a nanobiological system for the development of new products, and
- fortification of food by omega-3 fatty acids, haem, lycopene, beta-carotene, phytosterols, DHA/EPA, etc (Rashidi and Khosravi-Darani, 2011).

23.4 Process technology

The properties of nanomaterials are size and shape dependent, and thus the study of methods for their preparations is one of the primary research areas. Traditionally, the synthetic approaches to nanomaterials have been divided into two categories. A typical 'top-down' procedure is also called a physical method. It involves the mechanical grinding of bulk material and the subsequent stabilization of the resulting nanosized particles by the addition of colloidal protecting agents. A 'bottom-up' procedure attempts to build nanomaterials and devices one molecule/atom at a time, in much the same way that living organisms synthesize macromolecules (Singh, Kulkarni and Dash, 2013).

Several methods are available for the preparation of nanoparticles and allied materials. The overview in the form of a flowchart (Figure 23.2) shows some of these methods of preparation, which differ not only in the use of ingredients but also on the approach of preparation.

23.4.1 Top-down approach

The top-down approach involves physically reducing materials to nanometer size via such processes as grinding, milling, etching and lithography. The commercial scale production of nanomaterials today primarily involves the "top-down" approach. It is, however, believed that as nanotechnology develops there will be an increase in the use of the 'bottom-up' approach, where nanomaterials are built from individual atoms and molecules that have the capacity to self-assemble (Ofori and Hsieh, 2013; Sanguansri and Augustin, 2006).

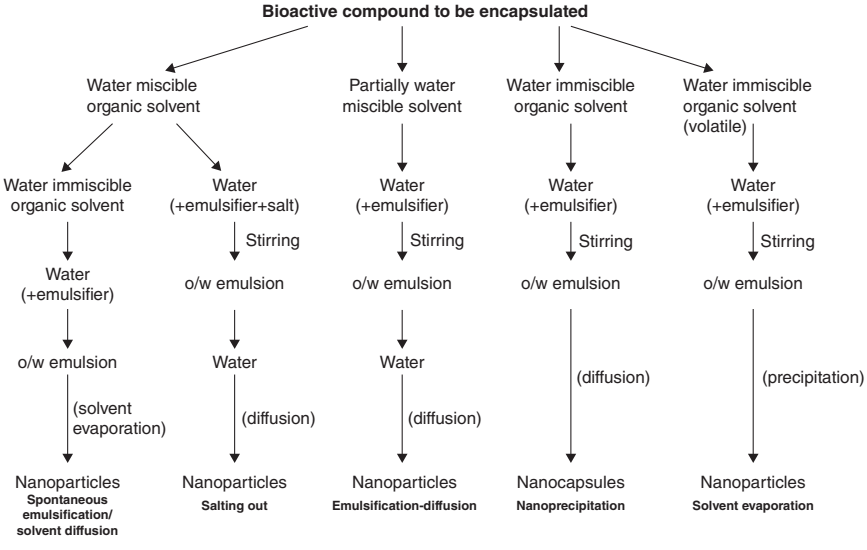

Figure 23.2 Overview of nanoparticle manufacture methods (source: Weiss *et al.* 2006. Reproduced with permission of John Wiley & Sons, Ltd.)

The "top-down" approach involves size reduction by the application of a force. The degree of control and refinement in size reduction processes affects the properties of the materials produced. Size usually relates to functional behaviour of food materials. A smaller size means a bigger surface area and is desirable for purposes such as improved water absorption, flavour release, bioavailability and faster rates of catalysis. Uniformity or a narrow size distribution is also required for better control of functionality and product quality. The three types of force used in the size reduction of foods are compression, impact and shear. Impact and shear are important in the formation of nanoparticles for food applications. The specific processes of nanoparticle preparation through the top-down approach are dry milling, high pressure homogenization and microfluidization, ultrasound emulsification, membrane emulsification, rapid expansion of supercritical fluids, electrified coaxial liquefied jets, etc (Sanguansri and Augustin, 2006).

Electrospinning is another method of preparing a nanomaterial through the top-down approach. It is gaining importance in recent years due to its advantages, such as increased efficiency, simple setup, versatility of nanomaterials produced and cost-effective procedure. This technique is especially used for preparing nanofibres whose diameter ranges between 10 and 1000 nm. The key advantage of producing fibres with extremely small diameters is their large surface-to-mass ratio, high porosity and superior mechanical performance. The other useful methods of preparation of nanofibres are drawing, phase separation, template synthesis and self-assembly (Kriegel *et al.*, 2008).

23.4.2 Bottom-up approach

The "bottom-up" approach relies on the self-assembling properties of molecules under thermodynamic control to build supramolecular structures, microstructures and higher hierarchical structures to produce functional materials. The forces between the building blocks can be influenced by a number of factors like temperature, concentration, pH and ionic strength of the system, the mechanical force (pressure, shear, extension and ultrasound) or the electric and magnetic field strengths. Examples of self-assembled nanostructures in food include the organization of the casein micelle, the structures formed in protein–polysaccharide coacervates and liposomes (Sanguansri and Augustin, 2006).

Self-assembling polymers and emulsification techniques are the two most utilized processes for the preparation of nanoparticles through the bottom-up approach. This approach allows easier control of the structure and properties of nanomaterials formed and conserves more energy compared to top-down synthesis. Nevertheless, each method has unique fabrications, which are continuously being refined to achieve the desired properties for distinct applications in foods and bioactives (Kuan *et al.*, 2012).

Apart from these methods for the production of nanoparticle, there are certain other methods of preparation of polymer nanoparticles. The term 'polymer nanoparticles' is a collective terminology given for any type of polymer nanoparticles, but specifically for nanospheres and nanocapsules. Nanospheres are matrix particles whose entire mass is solid and in which molecules may be adsorbed at the sphere surface or encapsulated within the particle. Nanocapsules are vesicular systems. They can act as a kind of reservoir, where the entrapped substances are confined to a cavity consisting of a liquid core (either oil or water) surrounded by a solid material shell (Rao and Geckeler, 2011).

23.5 Regulatory and safety issues

The potential for the use of nanotechnology in the agri-food sector is ambiguous about their use in the food sector because of suspected health risks and environmental concerns (Kumari and Yadav, 2013). Nanoparticles, due to unique characteristics, including their small size, can cross cell boundaries or pass directly from the lungs into the blood stream and ultimately reach various organs in the body. Hence, they may pose a higher risk than the same mass and material of larger particles (Kumari and Yadav, 2013).

23.5.1 Risks and safety issues

The risks involved in the consumption of a nanoscale food product should be assessed properly before commercialization. Risk assessment is a scientific procedure for estimating a risk and understanding the factors that influence it. Starting with the problem formulation, the process comprises four elements: hazard identification, exposure assessment, hazard characterization and risk characterization. Hazard identification consists of identifying known or potential adverse health effects in humans that are associated with exposure to a biological, physical or chemical agent. Exposure assessment involves the qualitative and/or quantitative evaluation of the likely intake of the agent via food. Risk characterization integrates hazard identification and characterization and exposure assessment into an estimation of the likely adverse effects (FAO/WHO, 2010).

Nanoparticles may pass the epithelial barrier lining of the digestive tract. After passage through the epithelium, either across cells or via endocytosis, nanoparticles can enter the capillaries and can appear in either the systemic circulation or the portal circulation to the liver. An important property of nanoparticles is their interaction with proteins. Protein adsorption to nanoparticles may enhance membrane crossing and cellular penetration. Furthermore, interaction with nanoparticles may affect the tertiary structure of a protein, resulting in malfunction. Hence, the potential of a nanoparticle to

disrupt the gastrointestinal barrier should be addressed during safety assessments (FAO/WHO, 2010).

Nanostructures are mostly used with relation to the field of food in the form of packaging materials but there are limited scientific data available about the migration of nanostructures from packaging materials into food (de Azeredo, 2013). Although bulk zinc oxide used as a nanocomposite is nontoxic, Sharma *et al.* (2009) demonstrated that ZnO nanoparticles have a genotoxic potential in epidermal cells. The migration of nanoparticles from the packaging material could also eventually promote sensory changes to foods. Titanium oxide, for instance, promotes lipid oxidation in cell membranes and could thus cause rancidity resulting from lipid oxidation in foods (de Azeredo, 2013; Jing *et al.*, 2011). Benn *et al.* (2010) recorded information on the migration of silver nanoparticles and quantified the migration of silver ions in consumer goods, finding that silver ions and nanoparticles migrate at levels that approximate the expected toxicity in some goods.

Another crucial factor that concerns the safety issues is the potential passage through natural barriers like the cellular barriers, blood-brain barrier, placental barrier and the blood-milk barrier by the nanomaterials. The permeability of the blood-brain barrier is highly restricted to molecules that are either lipophillic, actively transported or are small soluble molecules (<500 Da). This barrier may therefore represent a strict defense mechanism from blood-borne particle exposure that limits the distribution of nanoparticles to the brain. However, evidence exists that distribution to the brain might occur for some nanoparticles (Bouwmeester *et al.*, 2009).

23.5.2 Regulatory affairs

At present, there are no special regulations for using nanotechnology in foods. The Food and Drug Administration (FDA) regulates on a product-by-product basis. The technology-based regulations are not provided by the FDA (Weiss, Takhistov and McClements, 2006; FDA, 2004). FDA has traditionally regulated many products with particulate material in the nano-size range but has not focused on applied technology for their preparation (FDA, 2004).

Recommendations by the Royal Society and the Royal Academy of Engineering, commissioned by the UK government to assess the potential impact of nanotechnology, included a call for identification of the use of nanoparticles in ingredient lists. The UK government agreed that this was necessary for consumers to make informed decisions and that modifications to current labelling requirements would be however necessary. The Institute of Food Science and Technology (IFST) suggested that when nanoparticles are used as food additives, the conventional E-numbering system for labelling should be used along with the subscript 'n' (Rashidi and Khosravi-Darani, 2011; Weiss, Takhistov and McClements, 2006).

The European Commission intends to use existing food laws with respect to food products derived through nanotechnology, where applicable. The European Commission also plans to use a case-by-case approach to risk assessment (Weiss, Takhistov and McClements, 2006).

The Codex Alimentarius contains a set of standards for food products and its handling. The Codex Alimentarius was updated through the use of nanotechnology in food and agriculture. FAO and WHO have conducted an expert consultation in 2010 that identified the applications of nanotechnology in the food sector at present or in the future and the potential food safety issues, as well as exploring areas for future research and international guidance (Rashidi and Khosravi-Darani, 2011; FAO/WHO, 2010). It is anticipated that nanotechnology standards are being developed by organizations such as the International Standards for Organization (ISO) and ASTM International on terminology, nomenclature, measurement and characterization, environment, safety and health (Rashidi and Khosravi-Darani, 2011; ISO, 2010).

In Australia, like Europe, nanotechnologies are regulated by horizontal legislation. The NICNAS (National Industrial Chemicals Notification and Assessment Scheme) regulates chemicals for the protection of human health and the environment. They have recently introduced new administrative processes to address nanotechnology (NICNAS, 2010).

23.5.3 Precautionary measures

The important precautionary measure before ingesting a nanoparticle is to characterize it. Though there are certain analytical techniques available to characterize a nanoparticle like HPLC (high performance liquid chromatography), UPLC (ultra performance liquid chromatography), FFF (field flow fractionation) and capillary electrophoresis, *in situ* characterization of nanoparticles is essential (Magnuson, Jonaitis and Card, 2011; FAO/WHO, 2010).

A nonexhaustive list of equipment required to characterize nanoparticle includes: SEM (scanning electron microscope), TEM (transmission electron microscope), ESEM (environmental scanning electron microscope), FEG-ESEM (field emission gun-environmental scanning electron microscope), EDS (energy dispersive system), XRD (X-ray diffractometry) and dynamic light scattering (DLS). UV-Vis (ultraviolet-visible spectroscopy) can be used for the physical and chemical characterization of size, morphology, chemical composition and crystallinity (FAO/WHO, 2010).

The other precautionary step involves the study of the biokinetics of the nanoparticle in a human system. This whole cascade of events, which occurs following ingestion, determines the internal exposure of the organs to potentially toxic substances. Toxicological properties of the nanoparticles are important characteristics to understand the undesirable effects of the

nanoparticle. *In vitro* and *in vivo* testing should be performed and the optimum dosage levels should be determined for better utilization of the products. The clinical trials along with the dose-response relationship and determination of no-observed-effect level (NOEL) may also play a vital role in safe consumption. Exposure assessment involving the human exposure to nanomaterials as a component in the food matrix should also be characterized. Human contact of nanoparticles through oral, inhalatory and dermal routes should also be studied. Moreover, residual and disposal studies are also important (Cockburn *et al.*, 2012; Magnuson, Jonaitis and Card, 2011; FAO/WHO, 2010; Bouwmeester *et al.*, 2009).

23.6 Conclusions

The current level of nanotechnology applications in food and related sectors is still an emerging feature for most countries despite a steady increase in the number of published research articles and available products. The vast majority of new developments is still at R&D or the near-market stages. The lack of information on commercial activity in this area and estimates of the current and future market share of nanotechnology-enabled food products vary widely. The applications of nanotechnology in the food sector are extensively elaborated in the present chapter along with the methods of preparing nanoparticles; these are of interest for devising novel encapsulation materials and methods with an overall objective of improving the nutritional quality and stability of the functional ingredients. The applications of nanotechnology in food packaging seem to promise commercial success in the near future. The introduction of food products arising out of nanotechnology into the market place requires education of the public about nanotechnology to increase understanding of the technology and its implications for people.

References

Aboufazeli, F., Zhad, H.R.L.Z., Sadeghi, O., Karimi, M. and Najafi, E. (2013) Novel ion imprinted polymer magnetic mesoporous silica nano-particles for selective separation and determination of lead ions in food samples. *Food Chemistry*, **141**, 3459–3465.

Ahmad, A., Mukherjee, P., Senapati, S., Mandal, D., Khan, M.I., Kumar, R. and Sastry, M. (2003) Extracellular biosynthesis of silver nanoparticles using the fungus *Fusarium oxysporum*. *Colloids and Surfaces B – Biointerfaces*, **28**, 313–318.

Asmatulu, R. (ed.) (2013) *Nanotechnology Safety*. Elsevier, Oxford, England.

Astete, C.E., Sabliov, C.M., Watanabe, F. and Biris, A. (2009) Ca^{2+} cross-linked alginic acid nanoparticles for solubilization of lipophilic natural colorants. *Journal of Agricultural and Food Chemistry*, **57**, 7505–7512.

Augustin, M.A. and Oliver, C.M. (2012) An overview of the development and applications of nanoscale materials in the food industry. In: *Nanotechnology in the Food, Beverage and Nutraceutical Industries*, Huang, Q. (ed.). Woodhead Publishing Limited, Cambridge, England, pp. 1–39.

Avella, M., Avolio., R, Pace, E.D., Errico, M.E., Gentile, G. and Volpe, M.G. (2013) Polymer-based nanocomposites for food packaging applications. In: *Bio-Nanotechnology: A Revolution in Food, Biomedical and Health Sciences*, Bagchi, D., Bagchi, M., Moriyama, H. and Shahidi, F. (eds). John Wiley & Sons, Ltd, Chichester, West Sussex, England, pp. 212–226.

Baeumner, A. (2004) Nanosensors identify pathogens in food. *Food Technology*, **58**, 51–55.

Bagchi, D., Bagchi, M., Moriyama, H. and Shahidi, F. (eds) (2013) *Bio-Nanotechnology: A Revolution in Food, Biomedical and Health Sciences*. John Wiley & Sons, Ltd, Chichester, West Sussex, England.

Balcão, V.M., Costa, C.I., Matos, C.M., Moutinho, C.G., Amorim, M., Pintado, M.E., Gomes, A.P., Vila, M.M. and Teixeira, J.A. (2013) Nanoencapsulation of bovine lactoferrin for food and biopharmaceutical applications. *Food Hydrocolloids*, **32**, 425–431.

Benn, T., Cavanagh, B., Hristovski, K., Posner, J.D., and Westerhoff, P. (2010). The release of nanosilver from consumer products used in the home. *Journal of Environmental Quality*, **39**, 1875–1882.

Bouwmeester, H., Dekkers, S., Noordam, M.Y., Hagens, W.I., Bulder, A.S., de Heer, C., ten Voorde, S.E.C.G., Wijnhoven, S.W.P., Marvin, H.J.P. and Sips, A.J.A.M. (2009) Review of health safety aspects of nanotechnologies in food production. *Regulatory Toxicology and Pharmacology*, **53**, 52–62.

Burris, K.P. and Stewart, C.N. (2012) Fluorescent nanoparticles: sensing pathogens and toxins in foods and crops. *Trends in Food Science and Technology*, **28**, 143–152.

Chau, C.-F., Wu, S.-H. and Yen, G.-C. (2007) The development of regulations for food nanotechnology. *Trends in Food Science and Technology*, **18**, 269–280.

Chaudhry, Q. and Castle, L. (2011) Food applications of nanotechnologies: an overview of opportunities and challenges for developing countries. *Trends in Food Science and Technology*, **22**, 595–603.

Chaudhry, Q., Castle, L. and Watkins, R. (eds) (2010) *Nanotechnologies in Food*. The Royal Society of Chemistry, Cambridge, England.

Chaudhry, Q., Watkins, R. and Castle, L. (2010) Nanotechnologies in the food arena: new opportunities, new questions, new concerns. In: *Nanotechnologies in Food*, Chaudhry, Q., Castle, L. and Watkins, R. (ed.). The Royal Society of Chemistry, Cambridge, England, pp. 1–16.

Chen, H., Weiss, J. and Shahidi, F. (2006) Nanotechnology in nutraceuticals and functional foods. *Food Technology*, **60**, 30–36.

Cockburn, A., Bradford, R., Buck, N., Constable, A., Edwards, G., Haber, B., Hepburn, P., Howlett, J., Kampers, F., Klein, C., Radomski, M., Stamm, H., Wijnhoven, S. and Wildemann, T. (2012) Approaches to the safety assessment of engineered nanomaterials (ENM) in food. *Food and Chemical Toxicology*, **50**, 2224–2242.

Cushen, M., Kerry, J., Morris, M., Cruz-Romero, M. and Cummins, E. (2012) Nanotechnologies in the food industry – recent developments, risks and regulation. *Trends in Food Science and Technology*, **24**, 30–46.

de Azeredo, H.M.C. (2013) Antimicrobial nanostructures in food packaging. *Trends in Food Science and Technology*, **30**, 56–69.

del Carlo, M. and Compagnone, D. (2008) Recent advances in biosensor technology for food safety. *Agro Food Industry Hi-Tech*, **19**, 32–35.

Dhall, R.K. (2013) Advances in edible coatings for fresh fruits and vegetables: a review. *Critical Reviews in Food Science and Nutrition*, **53**, 435–450.

Donsì, F., Annunziata, M., Sessa, M. and Ferrari, G. (2011) Nanoencapsulation of essential oils to enhance their antimicrobial activity in foods. *LWT – Food Science and Technology*, **44**, 1908–1914.

Duncan, D.V. (2011) Applications of nanotechnology in food packaging and food safety: barrier materials, antimicrobials and sensors. *Journal of Colloid and Interface Science*, **363**, 1–24.

Falguera, V., Quintero, J.P., Jiménez, A., Muñoz, J.A. and Ibarz, A. (2011) Edible films and coatings: structures, active functions and trends in their use. *Trends in Food Science and Technology*, **22**, 292–303.

FAO/WHO (Food and Agriculture Organization of the United Nations/World Health Organization) (2010) FAO/WHO Expert Meeting on the Application of Nanotechnologies in the Food and Agriculture Sectors: Potential Food Safety Implications: Meeting Report, Rome, Italy, 130 pp.

Fathi, M., Mozafari, M.R. and Mohebbi, M. (2012) Nanoencapsulation of food ingredients using lipid based delivery systems. *Trends in Food Science and Technology*, **23**, 13–27.

FDA (Food and Drug Administration) (2004) FDA regulation of nanotechnology products. Available at: http: //www.fda.gov/nanotechnology/regulation.html.

Frewer, L.J., Norde, W., Fischer, A. and Kampers, F. (eds) (2011) *Nanotechnology in the Agri-Food Sector: Implications for the Future*. Wiley-VCH Verlag GmbH & Co., Weinheim, Germany.

Gökmen, V., Mogol, B.A., Lumaga, R.B., Fogliano, V., Kaplun, Z. and Shimoni, E. (2011) Development of functional bread containing nanoencapsulated omega-3 fatty acids. *Journal of Food Engineering*, **105**, 585–591.

Graveland-Bikker, J.F. and de Kruif, C.G. (2006) Unique milk protein based nanotubes: food and nanotechnology meet. *Trends in Food Science and Technology*, **17**, 196–203.

Gu, Y.S., Decker, A.E. and McClements, D.J. (2005) Production and characterization of oil-in-water emulsions containing droplets stabilized by multilayer membranes consisting of [beta]-lactoglobulin, [iota]-carrageenan and gelatin. *Langmuir*, **21**, 5752–5760.

Gutiérrez, F.J., Albillos, S.M., Casas-Sanz, E., Cruz, Z., García-Estrada, C., García-Guerra, A., García-Reverter, J., García-Suárez, M., Gatón, P., González-Ferrero, C., Olabarrieta, I., Olasagasti, M., Rainieri, S., Rivera-Patiño, D., Rojo, R., Romo-Hualde, A., Sáiz-Abajo, M.-J. and María-Luisa, M. (2013) Methods for the nanoencapsulation of β -carotene in the food sector. *Trends in Food Science and Technology*, **32**, 73–83.

Halder, D., Mitra, A., Bag, S., Raychaudhuri, R. and Chakraborty, R. (2011) Study on gelatin-silver nanoparticle composite towards the development of bio-based antimicrobial film. *Journal of Nanoscience and Nanotechnology*, **11**, 1–5.

Huang, Q. (ed.) (2012) *Nanotechnology in the Food, Beverage and Nutraceutical Industries*. Woodhead Publishing Limited, Cambridge, England.

Huang, Q., Given, P. and Qian, M. (2009) *Micro/Nano Encapsulation of Active Food Ingredients*, vol. 1007. Oxford University Press, Washington DC, USA.

Huang, Q., Yu, H. and Ru, Q. (2010) Bioavailability and delivery of nutraceuticals using nanotechnology. *Journal of Food Science*, **75**, R50–R57.

Imran, M., Revol-Junelles, A.-M., Martyn, A., Tehrany, E.A., Jacquot, M., Linder, M. and Desobry, S. (2010) Active food packaging evolution: transformation from micro- to nanotechnology. *Critical Reviews in Food Science and Nutrition*, **50**, 799–821.

Imran, M., Revol-Junelles, A.-M., René, N., Jamshidian, M., Akhtar, M.J., Tehrany, E.A., Jacquot, M. and Desobry, S. (2012) Microstructure and physico-chemical evaluation of nano-emulsion-based antimicrobial peptides embedded in bioactive packaging films. *Food Hydrocolloids*, **29**, 407–419.

ISO (International Organization for Standardization) (2010) Nanotechnologies – Vocabulary – Part 1: Core terms, ISO/DTS 80004-1, 14 January 2010. International Organization for Standardization, Geneva, Switzerland.

Ivanov, S.V. and Rashevskaya, T.A. (2011) The nanostructure's management is the basis for a functional fatty foods' production. *Procedia Food Science*, **1**, 24–31.

Jin, T. and Gurtler, J. B. (2011) Inactivation of *Salmonella* in liquid egg albumen by animicrobial bottle coatings infused with allyl isothiocyanate, nisin and zinc oxide nanoparticles. *Journal of Applied Microbiology*, **110**, 704–712.

Jing, Z., Guo, D., Wang, W., Zhang, S., Qi, W. and Ling, B. (2011) Comparative study of titania nanoparticles and nanotubes as antibacterial agents. *Solid State Sciences*, **13**, 1797–1803.

Joseph, T. and Morrison, M. (2006) Nanotechnology in agriculture and food. Nanoforum Report, Institute of Nanotechnology, UK.

Kalia, A. and Parshad, V.R. (2013) Novel trends to revolutionize preservation and packaging of fruits/fruit products: microbiological and nanotechnological perspectives. *Critical Reviews in Food Science and Nutrition*, DOI: 10.1080/10408398.2011.649315.

Kayaci, F. and Uyar, T. (2012). Encapsulation of vanillin/cyclodextrin inclusion complex in electrospun polyvinyl alcohol (PVA) nanowebs: prolonged shelf-life and high temperature stability of vanillin. *Food Chemistry*, **133**, 641–649.

Kriegel, C., Arrechi, A., Kit, K., McClements, D.J. and Weiss, J. (2008) Fabrication, functionalization, and application of electrospun biopolymer nanofibers. *Critical Reviews in Food Science and Nutrition*, **48**, 775–797.

Kuan, C.-Y., Yee-Fung, W., Yuen, K.-H. and Liong, M.-T. (2012) Nanotech: propensity in foods and bioactives. *Critical Reviews in Food Science and Nutrition*, **52**, 55–71.

Kumari, A. and Yadav, S.K. (2013) Nanotechnology in agri-food sector. *Critical Reviews in Food Science and Nutrition*. DOI: 10.1080/10408398.2011.621095.

Lee, J.Y., Wang, X.Y., Ruengruglikit, C. and Huang, Q.R. (2007) Nanotechnology in food materials research. In: *Food Materials Science: Principles and Practice*, Aguilera, J.M. and Lillford, P.J. (eds). Springer, New York, USA, pp. 123–144.

Li, J., Su, Q., Li, K.-Y., Sun, C.-F. and Zhang, W.-B. (2013) Rapid analysis of phthalates in beverage and alcoholic samples by multi-walled carbon nanotubes/silica reinforced hollow fibre-solid phase microextraction. *Food Chemistry*, **141**, 3714–3720.

Llorens, A., Lloret, E., Picouet, P.A., Trbojevich, R. and Fernandez, A. (2012) Metallic-based micro and nanocomposites in food contact materials and active food packaging. *Trends in Food Science and Technology*, **24**, 19–29.

Lopez-Rubio, A., Gavara, R. and Lagaron, J.M. (2006) Bioactive packaging: turning foods into healthier foods through biomaterials. *Trends in Food Science and Technology*, **17**, 567–575.

Magnuson, B.A., Jonaitis, T.S. and Card, J.W. (2011) A brief review of the occurrence, use, and safety of food-related nanomaterials. *Journal of Food Science*, **76**, R126–R133.

Malsch, I. and Emond, C. (2014) *Nanotechnology and Human Health*. CRC Press, Boca Raton, Florida, USA.

McClements, D.J., Decker, E.A., Park, Y. and Weiss, J. (2009) Structural design principles for delivery of bioactive components in nutraceuticals and functional foods. *Critical Reviews in Food Science and Nutrition*, **49**, 577–606.

Miyagawa, H., Misra, M., Drzal, L.T., and Mohanty, A.K. (2005) Novel biobased nanocomposites from functionalized vegetable oil and organically modified layered silicate clay. *Polymer*, **46**, 445–453.

Moraru, C.I., Panchapakesan, C.P., Huang, Q., Takhistov, P., Liu, S. and Kokini, J.L. (2003) Nanotechnology: a new frontier in food science. *Food Technology*, **57**, 24–29.

Moraru, C., Huang, Q.R., Takhistov, P., Dogan, H. and Kokini, J. (2009) Food nanotechnology: current developments and future prospects. In: *Global Issues in Food Science and Technology*, Barbosa-Cánovas, G.V., Mortimer, A., Colonna, P., Lineback, D., Spiess, W. and Buckle, K. (eds) Academic Press, Elsevier, New York, USA, pp 369–399.

Moura, M.R., Mattoso, L.H.C. and Zucolotto, V. (2012) Development of cellulose-based bactericidal nanocomposites containing silver nanoparticles and their use as active food packaging. *Journal of Food Engineering*, **109**, 520–524.

Nachay, K. (2007) Analyzing nanotechnology. *Food Technology*, **61**, 34–36.

Nanda, A. and Saravanan, M. (2009) Biosynthesis of silver nanoparticles from *Staphylococcus aureus* and its antimicrobial activity against MRSA and MRSE. *Nanomedicine – Nanotechnology Biology and Medicine*, **5**, 452–456.

National Nanotechnology Initiative (2006) Available from: http://www.nano.gov/html/facts/whatIsNano.html. Assessed in September 2013.

Nel, A., Xia, T., Madler, L. and Li, N. (2006) Toxic potential of materials at the nanolevel. *Science*, **311**, 622–627.

NICNAS (2010) National Industrial Chemicals Notification and Assessment Scheme. Australian Government Gazette. http://www.nicnas.gov.au/

Ofori, J.A. and Hsieh, Y.-H.P. (2013) Novel technologies for the production of functional foods. In: *Bio-Nanotechnology: A Revolution in Food, Biomedical and Health Sciences*, Bagchi, D., Bagchi, M., Moriyama, H. and Shahidi, F. (eds). John Wiley & Sons, Ltd, Chichester, West Sussex, England, pp. 143–162.

Padua, G.W. and Nonthanum, P. (2012) Material components for nanostructures. In: *Nanotechnology Research Methods for Foods and Bioproducts*, Padua, G.W. and Wang, Q. (eds). John Wiley & Sons, Inc., New Jersey, USA, pp. 5–18.

Padua, G.W. and Wang, Q. (eds) (2012) *Nanotechnology Research Methods for Foods and Bioproducts*. John Wiley & Sons, Inc., New Jersey, USA.

Park, H.M., Lee, W.K., Park, C.Y., Cho, W.J., and Ha, C.S. (2003) Environmentally friendly polymer hybrids. Part I: Mechanical, thermal, and barrier properties of thermoplastic starch/clay nanocomposites. *Journal of Materials Science*, **38**, 909–915.

Pegg, R.B. and Shahidi, F. (2007) Encapsulation, stabilization, and controlled release of food ingredients and bioactives. In: *Handbook of Food Preservation*, 2nd edn, Rahman, M.S. (ed.). CRC Press, Boca Raton, Florida, USA, pp. 509–568.

Rao, J.P. and Geckeler, K.E. (2011) Polymer nanoparticles: preparation techniques and size-control parameters. *Progress in Polymer Science*, **36**, 887–913.

Rashidi, L. and Khosravi-Darani, K. (2011) The applications of nanotechnology in food industry. *Critical Reviews in Food Science and Nutrition*, **51**, 723–730.

Rhim, J.-W. and Ng, P.K.W. (2007) Natural biopolymer-based nanocomposite films for packaging applications. *Critical Reviews in Food Science and Nutrition*, **47**, 411–433.

Sanguansri, P. and Augustin, M.A. (2006) Nanoscale materials development – a food industry perspective. *Trends in Food Science and Technology*, **17**, 547–556.

Sawicka, K., Gouma, P. and Simon, S. (2005) Electrospun biocomposite nanofibers for urea biosensing. *Sens Actuators*, **B108**, 585–588.

Sharma, V., Shukla, R.K., Saxena, N., Parmar, D., Das, M., and Dhawan, A. (2009) DNA damaging potential of zinc oxide nanoparticles in human epidermal cells. *Toxicology Letters*, **185**, 211–218.

Shibamoto, T., Kanazawa, K., Shahidi, F. and Ho, C.-T. (2008) Functional food and health: an overview. In: *Functional Food and Health*, Shibamoto, T., Kanazawa, K., Shahidi, F. and Ho, C.-T. (eds). ACS Symposium Series 993, Oxford University Press, Washington DC, USA, pp. 1–6.

Singh, S.K., Kulkarni, P.P. and Dash, D. (2013) Biomedical applications of nanomaterials: an overview. In: *Bio-Nanotechnology: A Revolution in Food, Biomedical and Health Sciences*, Bagchi, D., Bagchi, M., Moriyama, H. and Shahidi, F. (eds). John Wiley & Sons, Ltd, Chichester, West Sussex, England, pp. 3–32.

Song, X.-Y., Shi, Y.-P. and Chen, J. (2013) Carbon nanotubes-reinforced hollow fibre solid-phase microextraction coupled with high performance liquid chromatography for the determination of carbamate pesticides in apples. *Food Chemistry*, **139**, 246–252.

Sorrentino, A., Gorrasi, G. and Vittoria, V. (2007) Potential perspectives of bio-nanocomposites for food packaging applications. *Trends in Food Science and Technology*, **18**, 84–95.

Sozer, N. and Kokini, J.L. (2009) Nanotechnology and its applications in the food sector. *Trends in Biotechnology*, **27**, 82–89.

Vaclavik, V.A. and Christian, E.W. (2008) *Essentials of Food Science*. Springer, New York, USA.

Wang, G.-Y., Chen, J. and Shi, Y.-P. (2013) Preparation of microencapsulated xanthophyll for improving solubility and stability by nanoencapsulation. *Journal of Food Engineering*, **117**, 82–88.

Wang, Y., Zhang, Q., Zhang, C.-L. and Li, P. (2012) Characterisation and cooperative antimicrobial properties of chitosan/nano–ZnO composite nanofibrous membranes. *Food Chemistry*, **132**, 419–427.

Weiss, J., Takhistov, P. and McClements, D.J. (2006) Functional materials in food nanotechnology. *Journal of Food Science*, **71**, 107–116.

Xiao-e, L., Green, A.N.M., Haque, S.A., Mills, A. and Durrant, J.R. (2004) Light-driven oxygen scavenging by titania/polymer nanocomposite films. *Journal of Photochemistry and Photobiology A – Chemistry*, **162**, 253–259.

Yang, L. and Li, Y. (2005) Quantum dots as fluorescent labels for quantitative detection of *Salmonella typhimurium* in chicken carcass wash water. *Journal of Food Protection*, **68**, 1241–1245.

24

High Pressure Processing: Current Status

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24.1 Introduction

High pressure processing (HPP) of foods is a relatively novel nonthermal processing technology, in which high hydrostatic pressure, usually between 200 to 700 MPa, is applied to food items, the most common objective being a substantial decrease of microbial load. In a typical high pressure process, the food products are loaded into a high pressure vessel (commonly a thick steel cylinder), which is filled with a pressure transmitting fluid (usually water) and then pressurized by pumping additional fluid. The high pressure is held for a specific period of time after which the vessel is depressurized and the product is removed. Among the main advantages of high pressure processing is that the food can be processed after packaging, minimizing the risk of post-process contamination. Figure 24.1 shows a typical setup of a high pressure processing system. The main components are the steel cylinder with thick walls and the yolk, which are required to contain the high pressures achieved inside the cylinder.

High pressure processing technology has been extensively researched and has been shown to be effective in the inactivation of vegetative cells of bacteria and fungus, eukaryote parasites, and viruses. However, the processing parameters required to achieve a certain level of microbial inactivation greatly vary between different bacterial strains and food substrates. It has also been extensively shown that the technology is not effective by itself in inactivating microbial or fungal spores. In order to achieve effective spore inactivation

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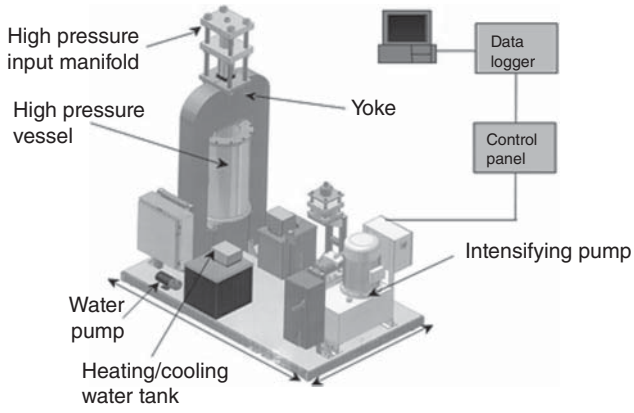

Figure 24.1 A typical high pressure vertical vessel with peripheral components (source: Reproduced with permission of Elmhurst Research, Inc)

levels, high pressure is usually combined with high temperature. In addition to the effects of high pressure on the microbial population in foods, there have been numerous studies on the effects of high pressure on enzyme activity, protein conformation, micronutrients and other bioactive compounds, colour and texture of foods, etc. Some reviews of high pressure processing technology have been carried out by Rendueles *et al.* (2011), Norton and Sun (2008), Doona and Feeherry (2007), Rastogi *et al.* (2007), Barbosa-Cánovas, Tapia and Cano (2005), Hendrickx and Knorr (2002), and Tauscher (1995).

This chapter will mainly focus on heat and mass transfer during high pressure processing, and briefly elucidate pressure nonuniformity in solid foods. Additionally, it will cover some of the literature on the effects of high pressure on bioactive compounds in foods and the possible mechanisms of microbial inactivation due to pressure. The reader is encouraged to review the references mentioned above for in-depth information on high pressure processing parameters for different foods and the effects on food components.

24.2 Heat transfer during high pressure processing

High pressure processing of foods is usually classified as a nonthermal technology; however, it is necessary to be applied in combination with other treatments, usually heating, in order to achieve meaningful inactivation of bacterial and fungal spores. For example, Sale, Gould and Hamilton (1970) carried out studies on spore suspensions of several species of *Bacillus* and achieved reductions no higher than 2 log CFU/mL after high pressure processing at 20 °C for 1 hour and pressures between 100 and

800 MPa. On the other hand, Ananta *et al.* (2001) were able to achieve reductions of 6 log CFU/g of *B. stearothermophilus* spores ATCC 7953 in mashed broccoli after a high pressure process at 600 MPa and 120 °C for 20 minutes.

The studies on the interaction between pressure and temperature in spores inactivation led to a petition to the FDA for the commercial use of pressure-assisted thermal sterilization (PATS) (Balasubramaniam, 2009). In the case of PATS, high pressure is only used as a means to reach the regular commercial sterilization temperatures, more rapidly and uniformly, through adiabatic compression heating. Farid (2006) filed a patent application for a process in which thermal expansion due to heating in a closed container is used to generate pressure and apply heat to a food product, and the interaction of both heat and pressure allows the temperature to be lower than if no pressure was being applied.

Since the thermal effects are important in high pressure processing, especially when it is aimed at the inactivation of spores, it is of interest to know and control the temperature at which high pressure processing takes place and any temperature gradients that may form during processing. Thermodynamics dictates that the temperature of most compressible substances will increase or decrease during compression or decompression, respectively. The temperature change when pressure is applied to a substance is given by

$$\frac{\mathrm{d}T}{\mathrm{d}P} = \frac{T\alpha_p}{\rho C_p} \tag{24.1}$$

where T is the temperature (K), P is the pressure (Pa), α_p is the thermal expansion coefficient (K^{-1}), ρ is the density (kg/m^3), and C_p is the isobaric heat capacity (J/kg K). The inconvenience with using this equation is that the properties are pressure and temperature dependent (Barbosa-Cánovas, Tapia and Cano, 2005). Figure 24.2 shows how density and heat capacity of water are pressure dependent, and is based on the NIST/ASME standard reference database 10 version 2.22 (Harvey, Peskin and Klein, 2010). Table 24.1 summarizes some of the findings of Buzrul *et al.* (2008) and Rasanayagam *et al.* (2003), who determined the temperature increase (adiabatic compression heating) of several liquids during high pressure processing. In both cases the initial temperature and pressure of the material, and the pressurization rate, were shown to influence the temperature increase rate.

A typical variation of pressure and temperature during a high pressure process is shown in Figure 24.3. Pressure increases during the pressurization stage, and so does the temperature, due to adiabatic compression heating. The pressure is then held constant for a specific amount of time in order to achieve the desired goal, followed by fast decompression. Although the pressure inside the vessel remains constant during the pressure hold time, the average

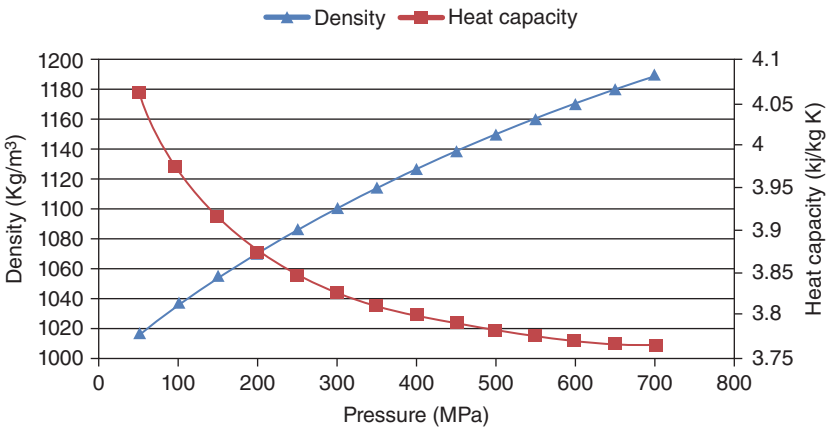

Figure 24.2 Density and heat capacity of pure water at 25 °C between 50 and 700 MPa (source: Data from Harvey *et al.* 2010)

Table 24.1 Temperature increase during high pressure processing of selected substances

Substance	Temperature increase (°C/100 MPa)
Water[1]	2.8
Orange juice[1]	2.8
Skim milk[1]	3.1
Salmon fish[2]	3 ± 0.1
Whole milk[1]	3.3
Ethylene glycol[1]	3.7
Crude beef fat[2]	4.4 ± 0.8
Propylene glycol[2]	5.1 ± 0.5
Soybean oil[2]	6.3 ± 0.4
Olive oil[2]	7.2 ± 0.2
Ethanol[1]	8.2

[1]From Buzrul *et al.* (2008).
[2]From Rasanayagam *et al.* (2003).

temperature normally decreases. As the temperature increases during pressurization due to adiabatic compression, a temperature gradient is established between the pressure transmitting medium and the colder vessel wall. The resulting heat transfer causes a decrease of temperature in the vessel contents during the pressure hold step and a lower final temperature compared to the initial value. Besides the heat transfer between the pressure transmitting medium and the vessel, there will also be temperature gradients between the different food components and between the food and the water, based on

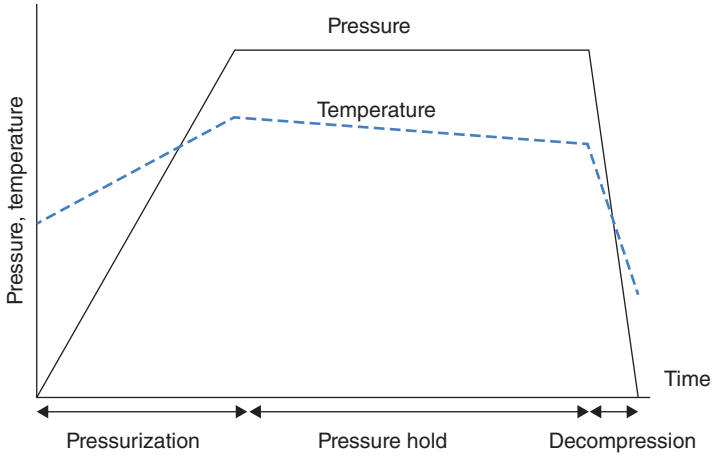

Figure 24.3 Variation of pressure and temperature during a typical high pressure process

each individual component's adiabatic compression heating value. Different components will reach different temperatures after pressurization, resulting in a complex and transient heat transfer process during the hold time. Experimental evidence has been obtained that suggested that the temperature gradients established in a high pressure vessel would generate variability in reaction kinetics for enzyme and microbial inactivation (Hartmann, Delgado and Szymczyk, 2003; Hartmann and Delgado, 2002, 2003; Denys, van Loey and Hendrickx, 2000). This indicates a potential use of certain enzymes as indicators of temperature uniformity in high pressure processing.

Several studies have been carried out aiming to model the temperature distribution during the pressure hold time. Hartmann (2002) carried out simulations on a 4 mL vessel and determined that the pressurization rate strongly influences the temperature gradients developed inside the vessel; for example, a maximum difference of 8 °C between two points inside the vessel at the end of the pressurization time was observed for a pressurization rate of 20 MPa/s while only a 6 °C difference was observed for a pressurization rate of 10 MPa/s, for an initial temperature of 21 °C and 500 MPa of pressure. He also determined that free convection would dominate the particle motions in the initial portion of the holding time, which was confirmed by Abdul Ghani and Farid (2007) by simulating the temperature distribution in water and in a mixture of beef fat pieces and water at 25 °C and 500 MPa of pressure. They also determined that the fat pieces would reach higher temperatures than the water, which is expected given that the adiabatic compression heating value of fat is higher than water. Pehl, Werner and Delgado (2002) studied the effect of viscosity on temperature gradients and determined that a solution of 50% sucrose would develop gradients six times higher than those of water due

to the higher viscosity. Khurana and Karwe (2009) conducted simulations of high pressure processing at different initial temperatures of the water and determined that the temperature gradients would be higher at higher initial temperatures. Khurana (2012) studied the effect of different positions for the water inlet used to pressurize the vessel and different vessel orientations; it was determined that the water inlet from the bottom would lead to more temperature uniformity in vertical vessels and that, in general, horizontal vessels would have a more uniform temperature distribution than vertical vessels.

A few studies have also been carried out aiming to reduce the temperature nonuniformity in the process. Knoerzer *et al.* (2007) studied the effect of a PTFE carrier inside the vessel for a process at 600 MPa and an initial temperature of 90 °C, and determined that its insulating effect with respect to the vessel walls would increase the temperature uniformity, allowing for 94.6% of the carrier volume to achieve a 12 log CFU/g reduction of *Clostridium botulinum* spores; without the carrier no reduction was achieved. In subsequent studies, Knoerzer *et al.* (2010) developed a software to optimize the wall thickness of a polymeric carrier in a high pressure unit, aiming to maximize the heat retention and uniformity, and Knoerzer, Buckow and Versteeg (2010) developed an approach to screen for insulating materials with adiabatic compression heating values different from water, which would allow better preservation of the temperature inside the carrier as the carrier walls would also heat during pressurization. Khurana (2012) simulated the effects of a 12.7 mm PTFE insulation layer between the vessel wall and the pressurizing medium in a 10 L vertical

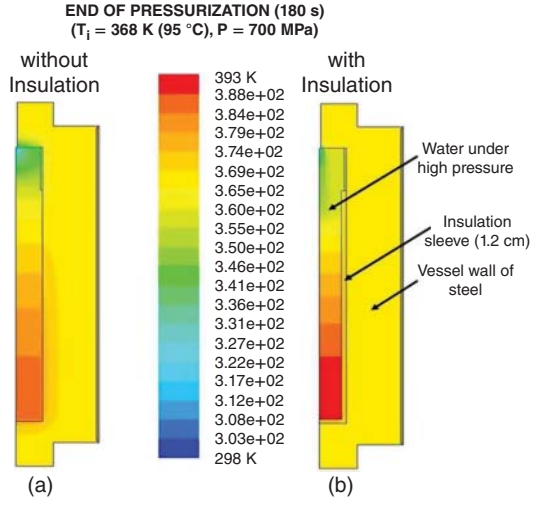

Figure 24.4 Isotherms in water and stainless steel vessel for vertical vessel at $T_i = 368$ K, $P = 700$ MPa, $T_{inlet} = 298$ K, $Q = 860\,000$ W/m^3 for ($0 \leq t = \leq 180$ s), (a) without insulation and (b) with insulation (12.7 mm thick) at the end of pressurization (180 s) (source: Khurana 2012. Reproduced with permission). *See plate section for colour version*.

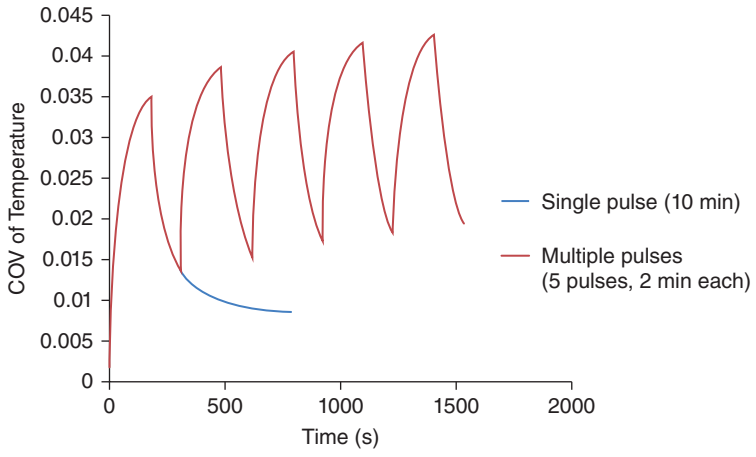

Figure 24.5 Coefficient of variance in a 10 L vertical vessel during high pressure processing at 700 MPa with initial temperature of 95 °C (source: Khurana 2012. Reproduced with permission)

vessel and in a 350 L horizontal vessel; the results for the vertical vessel are shown in Figure 24.4. A significant increase in temperature retention and uniformity was also observed in the horizontal vessel with an insulation layer.

Pressure cycling has also been investigated as a method to increase the lethality of the process. In pressure cycling, instead of having a hold time at a constant high pressure, the pressure chamber would be depressurized and pressurized again, with very short periods of pressure hold and almost no lag in between the cycles. Because the temperature after pressure release is lower than the initial temperature (see Figure 24.3), each cycle would start at a slightly lower temperature than the previous one. Pressure cycling has been found to be effective in enhancing microbial inactivation. Bradley *et al.* (2000), for example, found that pressure cycling enhanced the inactivation of lambda phage, a pathogen found in blood. Khurana (2012) investigated the temperature nonuniformities during pressure cycling and found that each cycle would have a slightly higher nonuniformity compared to the previous one, as shown in Figure 24.5. The temperature nonuniformity was expressed in terms of the coefficient of variation (COV).

24.3 Mass transfer during high pressure processing

High pressure-induced mass transfer has been explored as an enhancement over regular osmotic dehydration processes. Osmotic dehydration, which is a diffusion-based process, has been traditionally used in the food industry for partial removal of water from fruits and vegetables and for simultaneous

infusion of small solute molecules, such as sugar and salt, by immersing in concentrated solutions. Due to the difference in the internal and external osmotic pressures, water diffuses out from the vegetable into solution while solute molecules from the concentrated solution diffuse into the food matrix. This is usually a slow mass transfer process that can take several hours or even days. High pressure processing has been shown to disrupt the cell membranes of the substrate, which may allow a much faster mass transfer process and therefore dramatically reduce the time needed.

One of the earliest works on cell permeabilization using high pressure processing was done by Dornenburg and Knorr (1993), who studied the recovery of pigments from plant cells. Rastogi and Niranjan (1998) observed that high pressure processing would increase the diffusivity of water and sugar by a factor of four and two, respectively, compared to untreated samples of pineapples. Rastogi, Angerbach and Knorr (2000) observed similar effects in potato cylinders, this time in the diffusivity of NaCl. They reported the use of the cell permeabilization index (Z_p) to characterize the effect of high pressure on the cell membranes, as follows:

$$Z_p = \frac{\left(\frac{\sigma_h^i}{\sigma_h^t}\right)\sigma_l^t - \sigma_l^i}{\sigma_h^i - \sigma_l^i} \tag{24.2}$$

where σ is the electrical conductivity measured before processing (superscript i) or after processing (superscript t) and at a low frequency (subscript l) or high frequency (subscript h). The frequency values are dependent on the substrate. The value of Z_p varies between 0, for an intact cell system, and 1, for a completely disrupted cell system.

Additional work on enhancement of mass transfer with high pressure was done by Mahadevan and Karwe (2011), who observed that the infusion of quercetin into cranberries during high pressure processing was independent of the pressure applied between the range of 100 to 500 MPa and that Z_p alone was not a good predictor of an enhanced mass transfer coefficient, as high pressure would not increase the Z_p value for frozen–thawed cranberries and yet higher amounts of quercetin had been infused into these cranberries. Figures 24.6 and 24.7 show the infusion of quercetin and Z_p values measured in this study. Figure 24.8 shows the loss of cellular structure after high pressure processing of frozen–thawed cranberries.

The enhancement of mass transfer due to high pressure processing, at this point, has not attracted nearly as much attention as microbial inactivation, either by pressure alone or by the combination of pressure and heat. All the studies published so far point out that the rate of mass transfer is greatly increased and that this increase is dependent on the substrate and the solute. So far there is no general agreement on the mechanism by which high pressure enhances mass transfer, so multiple mechanisms are being hypothesized.

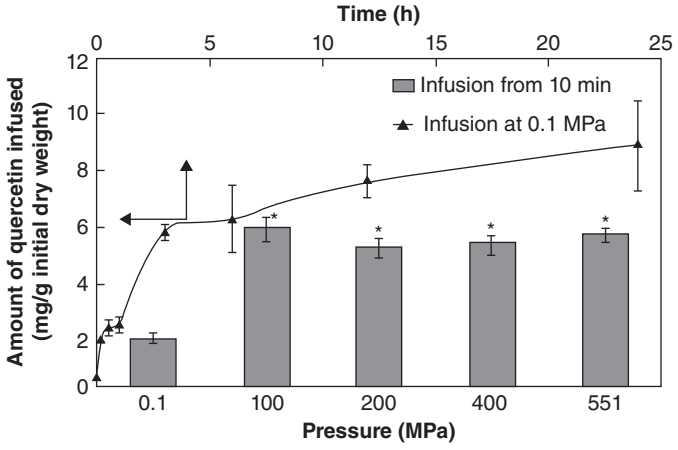

Figure 24.6 Quercetin infused in frozen–thawed cranberries at atmospheric conditions and during high pressure processing (source: Mahadevan and Karwe, 2011)

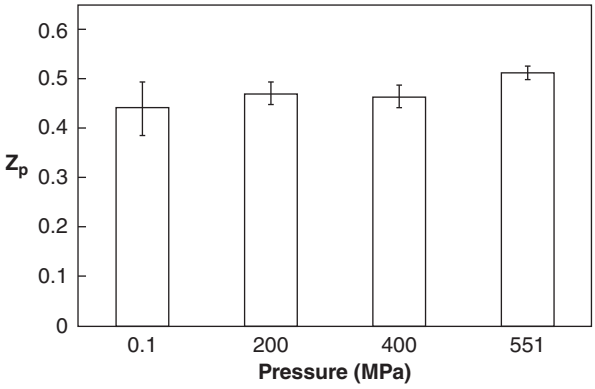

Figure 24.7 Z_p values of frozen–thawed cranberries before and after high pressure processing (source: Mahadevan and Karwe, 2011)

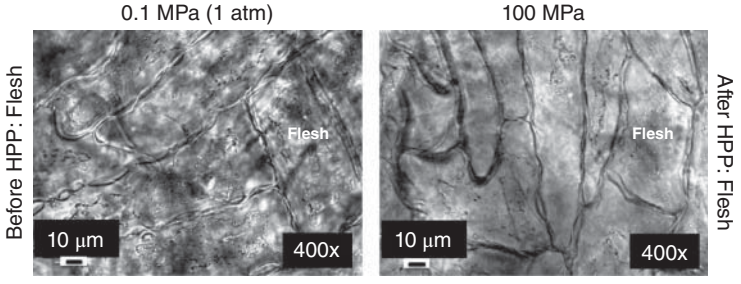

Figure 24.8 Microstructure of frozen–thawed cranberry flesh before (left) and after (right) high pressure processing (source: Mahadevan and Karwe, 2011)

24.4 Studies on nonuniformity of pressure in solid foods

In most of the currently published literature on high pressure processing of foods, the pressure is considered to be transmitted uniformly and quasi-instantaneously throughout the sample, regardless of size, shape or packaging (Rendueles *et al.*, 2011; Doona and Feeherry, 2007; Brennan, 2006; Barbosa-Cánovas, Tapia and Cano, 2005; Sun, 2005) and with no distinction between liquid and solid materials or homogeneous and heterogeneous foods. The only study suggesting the possibility of pressure not being uniform in solid foods was done by Minerich and Labuza (2003), who used the compression of a copper powder tablet during high pressure processing as an indicator of pressure and determined that the pressure at the center of a cooked ham piece, where a tablet had been inserted, was on average 9 MPa less than the pressurizing medium when pressures between 400 and 600 MPa were applied. Hartmann and Delgado (2004) and Hartmann, Mathmann and Delgado (2006) carried out numerical simulations of the application of hydrostatic pressure on yeast cells using known mechanical properties of the organelles and concluded that the pressure transmitted to the cytoplasm of the cell would be lower than the applied pressure at the cell wall, and that von Mises (shear) stress would develop in the cell wall, as shown in Figure 24.9.

From the point of view of solid mechanics, when hydrostatic pressure is applied to any material, including food items, the material will get compressed. The relationship between the applied pressure and the compression of a given material is characterized in terms of the bulk modulus (K), defined as

$$K = -V\frac{\partial P}{\partial V} \tag{24.3}$$

where P is the applied pressure and V is the volume of the material.

If a food item consists of two or more materials, such as a bone surrounded by muscle fibers, each component would be compressed differently depending on their individual bulk modulus. At the interface between the soft and the hard materials, the deformation of both will be limited by the deformation of the hard material due to friction or other attachments. This will cause a deformation in the soft material, similar to Figure 24.10.

The shape change, initiated at the interface of the solids, would cause shear strain, which in turn would cause the development of shear stresses, given by

$$\tau = G\gamma \tag{24.4}$$

where τ is the shear stress, G is the shear modulus of the soft material, and γ is the shear deformation.

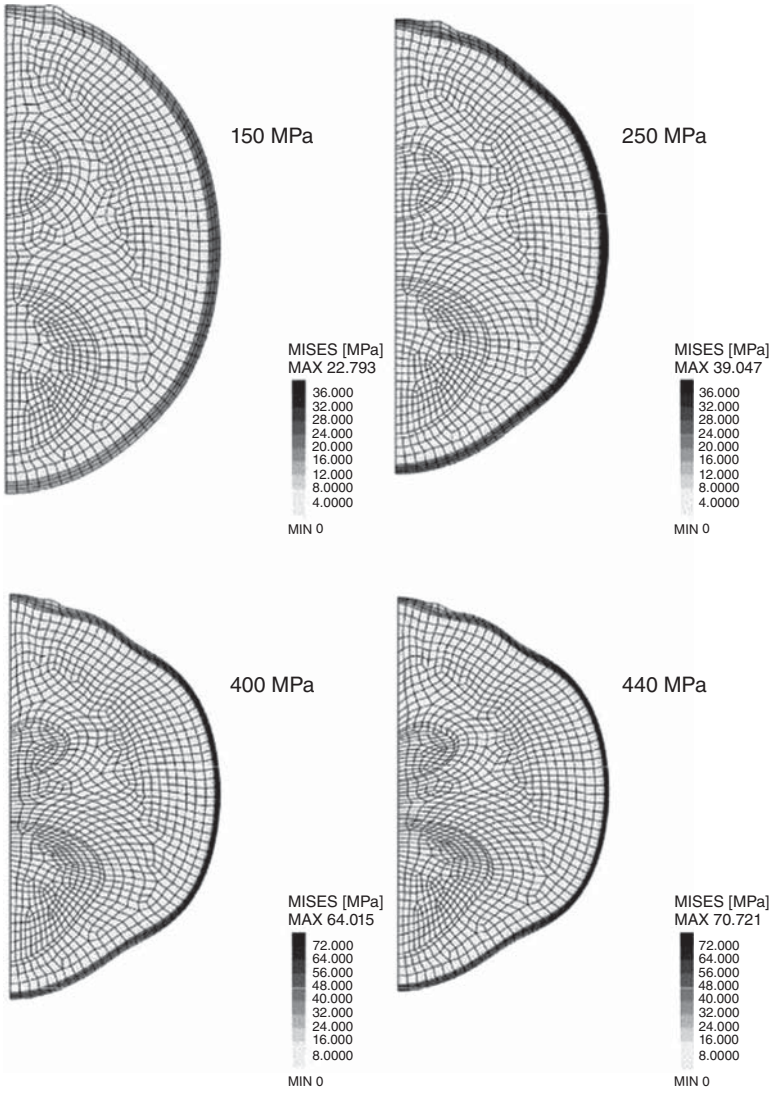

Figure 24.9 Von Mises stress developed in the cell wall of yeast cells when high hydrostatic pressure is applied (source: Hartmann *et al.*, 2005. Reproduced with permission of Elsevier)

Due to conservation of energy, the energy used to develop the shear stress would cause a decrease of pressure. A schematic of the pressure and shear profiles that would develop in this scenario are shown in Figure 24.11.

Currently, research is being carried out on nonuniformity of pressure in heterogeneous solids such as meats containing bones (Karwe, Schaffner and Cuitiño, 2012). This work is necessary if high pressure processing is to become

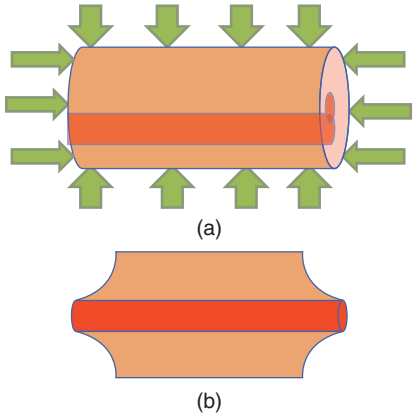

Figure 24.10 Soft cylindrical solid with a hard inclusion at the center (a) before high pressure is applied and (b) at high hydrostatic pressure. The combination of the surface pressure and the friction at the interface with the hard inclusion produces a deformation in the soft material

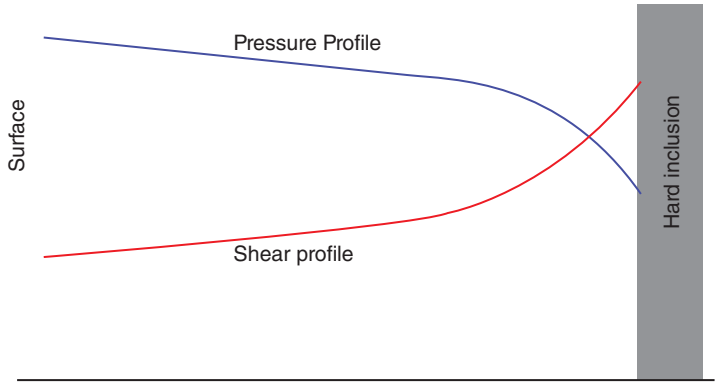

Figure 24.11 Schematic of pressure and shear distributions in a soft solid with a hard inclusion when high hydrostatic pressure is applied at the surface

more commonly used and applied on foods with inclusions or particulate foods, in order to ensure that the required microbial reduction is achieved at every location within the food product.

24.5 Effect of high pressure on bioactive compounds in foods

High pressure processing is promoted as a nonthermal technology that can better preserve the fresh-like aspects of food products compared to regular thermal processing; therefore it has been of interest to determine whether

food bioactive compounds, especially those found in fruits, are retained after processing. This topic has attracted the attention of numerous researchers, and several studies and review papers (Rawson *et al.*, 2011; Oey *et al.*, 2008a, 2008) have been published on this topic.

In general, high pressure processing is considered to have little effect on covalent bonds (Rastogi *et al.*, 2007). Nonetheless, it influences the equilibrium of a reaction by favoring the direction in which the volume is reduced, either toward the products for reactions with negative reaction volumes, that is, when the volume of the products is lower than the volume of the reactants, or towards the reactants for reactions with a positive reaction volume. Klärner *et al.* (1998) identified several cyclization reactions of 1-alkenes with negative reaction volumes; these could potentially affect the retention of bioactive compounds in fruits. De Ancos, Gonzales and Cano (2000) studied the carotenoid profile of persimmon fruits after high pressure and found significantly less degradation compared to thermal processing. In some specific pigments they observed an increase, and hypothesized that it could be due to disruption of cellular organelles or modification of the proteins that bound the carotenoids, which could enhance the release. Doblado, Frías and Vidal-Valverde (2007) measured the vitamin C content and trolox-equivalent antioxidant capacity of high pressure processed germinated cowpeas and observed a decrease in both as the pressure was increased. Unlike these two studies, Fernández García *et al.* (2001) observed no decrease in antioxidant capacity, vitamin C or carotene content in orange, lemon and carrot juice after high pressure processing.

Some insights on nutrient loss were obtained by Yen and Lin (1996). They measured vitamin C retention in thermally processed and high pressure processed guava purees and observed a lower decrease right after processing for high-pressur processed puree but faster degradation during storage. This suggests that high pressure retained vitamin C better than thermal processing, but at the same time the lower enzyme inactivation levels could have contributed to vitamin C loss during storage. Thakkar (2012) measured the oxygen radical absorbance capacity (ORAC) of the antioxidant, total phenolics and ellagic acid contents of Muscadine grape juice after thermal pasteurization and a microbiologically equivalent high pressure processing and found no difference between the treatments or with the unprocessed sample. After 8 weeks of storage at ambient temperature the ellagic content in the high pressure processed sample had increased, possibly due to hydrolysis of ellagitannins by enzymes not inactivated during processing.

The effects of thermal and high pressure processing on the antimutagenic activity of plant compounds have also been of interest. Butz *et al.* (1997) screened 14 fruits and vegetables using the Ames test and IQ carcinogen, and divided them into three groups: (1) resistant to both heat and pressure, like grapefruit and strawberry; (2) resistant to pressure but not heat, like carrots, cauliflower, kohlrabi, leek, and spinach; and (3) sensitive only to very

high pressures, like beet and tomatoes. The effects of fermentation and high pressure extraction on the antimutagenic activity of deodeok and Korean barberry were studied (Lee, He and Ahn, 2010; He *et al.*, 2010). It was determined that high pressure extraction achieved the highest antimutagenic activities for fermented samples.

It is evident that more research is needed on this topic, especially if 'healthier product' claims are desired for high pressure processed products for which consumers would be willing to pay a premium.

24.6 Mechanisms of microbial inactivation during high pressure processing

As mentioned before, numerous studies have been done to quantify the inactivation of different bacterial species in foods, using different pressure, time, and temperature combinations. On the other hand, there are very few studies dealing with the mechanism by which high pressure inactivates microorganisms. One of the main reasons for this is the difficulty of carrying out *in situ* studies, that is, studying microbial cells in real time while they are being pressurized or depressurized, and not just before and after processing. The application of high pressure to cells triggers a series of events in the cells, not all of them necessarily lethal. Among the events studied, membrane damage has been observed in numerous studies and it has been suggested that this is an important trigger of cell death during high pressure processing (Michiels, Bartlett and Aertsen, 2008).

Perrier-Cornet, Marechal and Gervais (1995) observed a permanent decrease by 10% of the volume of yeast cells after depressurization in an HPP cycle at 250 MPa for 15 min. Benito *et al.* (1999) studied the pressure resistance of different strains of *E. coli* 0157 H7 isolated from different outbreaks and were able to determine that the more pressure-sensitive strains also absorbed a fluorescent stain at a faster rate compared to more pressure-resistant strains after high pressure, suggesting that the susceptible strains had sustained more membrane damage. Additionally, the study indicated that the strains more resistant to pressure were also more resistant to acid, oxidative, and osmotic stresses. In another study, Perrier-Cornet, Hayert and Gervais (1999) were able to differentiate the inactivated cells from the surviving cells after high pressure processing and found a much larger volume reduction after decompression in the inactivated cells (35% volume loss) than in the surviving cells (10% volume loss). They also observed leakage of sodium, glycerol, calcium, and potassium ions from the cells to the medium. Manas and Mackey (2004) observed a leakage of proteins and RNA after application of 200 MPa of pressure.

Hartmann and Delgado (2004) carried out numerical simulations of stress distribution of yeast cells during HPP and concluded that Von Mises (shear) stress would develop at the cell wall and possibly disrupt it, but in the interior of the cell the stresses would be mostly hydrostatic; the effect on cell membrane was not included in this study. Although they did not experimentally verify their results, they found agreement with the experimental results obtained by Perrier-Cornet, Hayert and Gervais (1999). In a relatively recent study, Black *et al.* (2007) determined that the minerals and ions associated with the casein micelles (calcium, magnesium, citrate, and phosphate) increased the pressure resistance of *Listeria innocua* and hypothesized that the buffering capacity from phosphate and citrate ions, and membrane protection from calcium and magnesium would be the cause of this.

It has been observed that environmental conditions can affect the physical properties of the cell walls of bacteria, which could relate to their resistance to pressure treatments. Thwaites and Mendelson (1985) and Thwaites and Surana (1991) studied the effect of different relative humidity levels on the cell wall of *Bacillus subtilis* and observed that Young's modulus and tensile strength of the cell wall decreased as the relative humidity increased; that is, at dryer conditions, the cell walls were less susceptible to be deformed or broken. Similar conclusions were reached by Nikiyan, Vasilchenko and Deryabin (2010) for *Bacillus cereus* and *E. coli*, especially at RH levels below 84% for *E. coli* and below 65% for *Bacillus cereus*. This could explain the extreme resistance to pressure of several *Salmonella* strains in peanut butter, a very low moisture food, even though they were sensitive to pressure in peptone water suspensions (D'Souza, Karwe and Schaffner, 2012). In addition to cell wall strengthening, dehydration has also been shown to hinder the denaturation of proteins due to high pressure (Oliveira *et al.*, 1994), which could also explain the pressure resistance of *Salmonella* in peanut butter.

Besides the physical rupture of the membrane, the other changes that have been observed are permeabilization (that is, increased transfer of material from the cytoplasm to the medium) and inactivation of the F_0F_1 proton translocating ATPase (Michiels, Bartlett and Aertsen, 2008). A review on the available literature regarding the effects of high pressure on biological molecules was published by Cheftel (1995); ATPase inactivation, macromolecular transconformations, ionic dissociation and pH changes in water, changes in melting point and crystal structure were listed among the mechanisms studied that could be related to cell death. Pressure has also been observed to cause dissociation of ribosomes (reversible at low pressures and irreversible at high pressures), inactivation of RNA polymerase and DNA gyrase (an enzyme involved in DNA replication), and condensation of the nucleotide (Michiels, Bartlett and Aertsen, 2008). In a study with *Leuconostoc mesenteroides*, Kaletunç *et al.* (2004) determined that ribosomal

denaturation was the main cause of cell death, but also observed the formation of blisters in the surface of cells, suggesting additional membrane damage. Mentré and Hoa (2001) pointed out that high pressure has a stabilizing effect on DNA hydrogen bonds; this could hinder DNA transcription and therefore block the synthesis of vital proteins for the cell. Pope *et al.* (1975) measured the pressure sensitivity of protein synthesis in cells and found them to be different between *E. coli*, *Pseudomonas fluorescens*, and *Pseudomonas bathycetes*. They also concluded that the sensitivity changed with temperature. Wong and Heremans (1988) observed irreversible changes in the structure of chymotrypsinogen enzyme, due the reduction of α-helix and β-sheet substructures and the increase of random coil and turn conformations. They also observed that the pressure at which the denaturation started was higher when the pressurization rate was decreased.

The published studies on high pressure effects on bacterial spores have also not given a clear reason as to why they are so resistant to pressure (Reineke, Mathys and Knorr, 2011). Nguyen Thi Minh *et al.* (2011) studied the effects of sporulation of *Bacillus subtilis* at different conditions, such as temperature, pH, and calcium content, and determined that they influenced the resistance of the spores to both high temperature and pressure. A similar study was done by Olivier, Bull and Chapman (2012) with other *Bacillus* species; however, they found a negative correlation between developed temperature and pressure resistance at different sporulation temperatures, and found no trend between added minerals and resistance to pressure. Reineke, Mathys and Knorr (2011), also working with *Bacillus subtilis*, were able to determine that the inactivation of spores during high pressure and temperature treatments first triggered the spore germination before inactivation took place.

It is clear that more research is needed on this topic, especially if high pressure processing is to be used to achieve commercial sterility in low-acid foods. Fortunately, the research on pressure tolerance of microorganisms to high pressure processing is also of interest to other disciplines, such as marine biology and astrobiology (Sharma, 2011; Moeller *et al.*, 2008; Nicholson *et al.*, 2000), from which food science could benefit.

References

Abdul Ghani, A.G. and Farid, M.M. (2007) Numerical simulation of solid–liquid food mixture in a high pressure processing unit using computational fluid dynamics. *Journal of Food Engineering*, **80**, 1031–1042.

Ananta, E., Heinz, V., Schlüter, O. and Knorr, D. (2001) Kinetic studies on high-pressure inactivation of *Bacillus stearothermophilus* spores suspended in food matrices. *Innovative Food Science and Emerging Technologies*, **2**, 261–272.

Balasubramaniam, V.M.B. (2009) Opportunities and challenges in pressure-assisted thermal sterilization of low-ccid foods. In: *International Forum on Emerging Technologies in Food Processing*, University of Illinois (Urbana-Champaign), USA.

Barbosa-Cánovas, G.V., Tapia, M.A.S. and Cano, M.P. (2005) *Novel Food Processing Technologies*. CRC Press, Boca Raton, Florida, USA.

Benito, A., Ventoura, G., Casadei, M., Robinson, T. and Mackey, B. (1999) Variation in resistance of natural isolates of *Escherichia coli* O157 to high hydrostatic pressure, mild heat, and other stresses. *Applied and Environmental Microbiology*, **65**, 1564–1569.

Black, E.P., Huppertz, T., Fitzgerald, G.F. and Kelly, A.L. (2007) Baroprotection of vegetative bacteria by milk constituents: a study of *Listeria innocua*. *International Dairy Journal*, **17**, 104–110.

Bradley, D.W., Hess, R.A., Tao, F., Sciaba-Lentz, L., Remaley, A.T., Laugharn Jr, J.A. and Manak, M. (2000) Pressure cycling technology: a novel approach to virus inactivation in plasma. *Transfusion*, **40**, 193–200.

Brennan, J.G. (2006) *Food Processing Handbook*. Wiley-VCH, Weinheim, Germany.

Butz, P., Edenharder, R., Fister, H. and Tauscher, B. (1997) The influence of high pressure processing on antimutagenic activities of fruit and vegetable juices. *Food Research International*, **30**, 287–291.

Buzrul, S., Alpas, H., Largeteau, A., Bozoglu, F. and Demazeau, G. (2008) Compression heating of selected pressure transmitting fluids and liquid foods during high hydrostatic pressure treatment. *Journal of Food Engineering*, **85**, 466–472.

Cheftel, J.C. (1995) Review: high-pressure, microbial inactivation and food preservation (Revisión: Alta-presión, inactivación microbiológica y conservación de alimentos). *Food Science and Technology International*, **1**, 75–90.

de Ancos, B., Gonzalez, E. and Cano, M.P. (2000) Effect of high-pressure treatment on the carotenoid composition and the radical scavenging activity of persimmon fruit purees. *Journal of Agricultural and Food Chemistry*, **48**, 3542–3548.

Denys, S., van Loey, A.M. and Hendrickx, M.E. (2000) A modeling approach for evaluating process uniformity during batch high hydrostatic pressure processing: combination of a numerical heat transfer model and enzyme inactivation kinetics. *Innovative Food Science and Emerging Technologies*, **1**, 5–19.

Doblado, R., Frías, J. and Vidal-Valverde, C. (2007) Changes in vitamin C content and antioxidant capacity of raw and germinated cowpea (*Vigna sinensis* var. carilla) seeds induced by high pressure treatment. *Food Chemistry*, **101**, 918–923.

Doona, C.J. and Feeherry, F.E. (2007) *High Pressure Processing of Foods*. Blackwell Publishing, IFT Press, Ames, Iowa, Chicago, USA.

Dornenburg, H. and Knorr, D. (1993) Cellular permeabilization of cultured plant-tissues by high electric-field pulses or ultra high-pressure for the recovery if secondary metabolites. *Food Biotechnology*, **7**, 35–48.

D'souza, T., Karwe, M. and Schaffner, D.W. (2012) Effect of high hydrostatic pressure and pressure cycling on a pathogenic *Salmonella enterica* serovar cocktail inoculated into creamy peanut butter. *Journal of Food Protection*, **75**, 169–173.

Farid, M.M. (2006) *Pressure Assisted Thermal Sterilisation or Pasteurisation Method and Apparatus*. US Patent Application 12/296,789.

Fernández García, A., Butz, P., Bognàr, A. and Tauscher, B. (2001) Antioxidative capacity, nutrient content and sensory quality of orange juice and an orange–lemon–carrot juice product after high pressure treatment and storage in different packaging. *European Food Research and Technology*, **213**, 290–296.

Hartmann, C. (2002) Numerical simulation of thermodynamic and fluid-dynamic processes during the high-pressure treatment of fluid food systems. *Innovative Food Science and Emerging Technologies*, **3**, 11–18.

Hartmann, C. and Delgado, A. (2002) Numerical simulation of convective and diffusive transport effects on a high-pressure-induced inactivation process. *Biotechnology and Bioengineering*, **79**, 94–104.

Hartmann, C. and Delgado, A. (2003) The influence of transport phenomena during high-pressure processing of packed food on the uniformity of enzyme inactivation. *Biotechnology and Bioengineering*, **82**, 725–735.

Hartmann, C. and Delgado, A. (2004) Numerical simulation of the mechanics of a yeast cell under high hydrostatic pressure. *Journal of Biomechanics*, **37**, 977–987.

Hartmann, C., Delgado, A. and Szymczyk, J. (2003) Convective and diffusive transport effects in a high pressure induced inactivation process of packed food. *Journal of Food Engineering*, **59**, 33–44.

Hartmann, C., Mathmann, K. and Delgado, A. (2006) Mechanical stresses in cellular structures under high hydrostatic pressure. *Innovative Food Science and Emerging Technologies*, **7**, 1–12.

Harvey, A.H., Peskin, A.P. and Klein, S.A. (2010) NIST/ASME Steam Properties Database, 2.2 edn, National Institute of Standards and Technology.

He, X., Kim, S.S., Park, S.J., Seong, D.H., Yoon, W.B., Lee, H.Y., Park, D.S. and Ahn, J. (2010) Combined effects of probiotic fermentation and high-pressure extraction on the antioxidant, antimicrobial, and antimutagenic activities of deodeok (*Codonopsis lanceolata*). *Journal of Agricultural and Food Chemistry*, **58**, 1719–1725.

Hendrickx, M.E.G. and Knorr, D.W. (2002) *Ultra High Pressure Treatments of Foods*. Kluwer Academic/Plenum Publishers, New York, USA.

Kaletunç, G., Lee, J., Alpas, H. and Bozoglu, F. (2004) Evaluation of structural changes induced by high hydrostatic pressure in *Leuconostoc mesenteroides*. *Applied and Environmental Microbiology*, **70**, 1116–1122.

Karwe, M.V., Schaffner, D.W. and Cuitiño, A. (2012) Process non-uniformity during high hydrostatic pressure processing of heterogeneous foods. IFT Annual Meeting, 2012, Las Vegas, USA.

Khurana, M. (2012) Temperature distribution in vertical and horizontal high pressure vessels and its impact on food safety. PhD Thesis, Rutgers University, New Brunswick, New Jersey,USA.

Khurana, M. and Karwe, M.V. (2009) Numerical prediction of temperature distribution and measurement of temperature in a high hydrostatic pressure food processor. *Food and Bioprocess Technology*, **2**, 279–290.

Klärner, F.-G., Diedrich, M.K., Dierkes, G. and Gehrke, J.-S. (1998) Organic reactions at high pressure. the effect of pressure on cyclizations and homolytic bond cleavage. In: ISAACS, N.S. (ed.), *High Pressure Food Science, Bioscience and Chemistry*. The Royal Society of Chemistry, Cambridge, England.

Knoerzer, K., Buckow, R. and Versteeg, C. (2010) Adiabatic compression heating coefficients for high-pressure processing – a study of some insulating polymer materials. *Journal of Food Engineering*, **98**, 110–119.

Knoerzer, K., Juliano, P., Gladman, S., Versteeg, C. and Fryer, P.J. (2007) A computational model for temperature and sterility distributions in a pilot-scale high-pressure high-temperature process. *AIChE Journal*, **53**, 2996–3010.

Knoerzer, K., Buckow, R., Chapman, B., Juliano, P. and Versteeg, C. (2010) Carrier optimisation in a pilot-scale high pressure sterilisation plant – an iterative CFD approach

employing an integrated temperature distributor (ITD). *Journal of Food Engineering*, **97**, 199–207.

Lee, H.Y., He, X. and Ahn, J. (2010) Enhancement of antimicrobial and antimutagenic activities of Korean barberry (*Berberis koreana* Palib.) by the combined process of high-pressure extraction with probiotic fermentation. *Journal of Science of Food Agriculture*, **90**, 2399–2404.

Mahadevan, S. and Karwe, M.V. (2011) Enhanced infusion under high pressure: new insights. In: *International Congress on Engineering and Food,* Athens, Greece.

Manas, P. and Mackey, B.M. (2004) Morphological and physiological changes induced by high hydrostatic pressure in exponential- and stationary-phase cells of *Escherichia coli*: relationship with cell death. *Applied Environmental Microbiology*, **70**, 1545–1554.

Mentré, P. and Hoa, G.H.B. (2001) Effects of high hydrostatic pressures on living cells: a consequence of the properties of macromolecules and macromolecule-associated water. In: *International Review of Cytology*. Academic Press.

Michiels, C., Bartlett, D.H. and Aertsen, A. (2008) *High-Pressure Microbiology*. ASM Press, Washington, DC, USA.

Minerich, P.L. and Labuza, T.P. (2003) Development of a pressure indicator for high hydrostatic pressure processing of foods. *Innovative Food Science and Emerging Technologies*, **4**, 235–243.

Moeller, R., Horneck, G., Rabbow, E., Reitz, G., Meyer, C., Hornemann, U. and Stöffler, D. (2008) Role of DNA protection and repair in resistance of *Bacillus subtilis* spores to ultrahigh shock pressures simulating hypervelocity impacts. *Applied and Environmental Microbiology*, **74**, 6682–6689.

Nguyen Thi Minh, H., Durand, A., Loison, P., Perrier-Cornet, J.M. and Gervais, P. (2011) Effect of sporulation conditions on the resistance of *Bacillus subtilis* spores to heat and high pressure. *Applied Microbiology and Biotechnology*, **90**, 1409–1417.

Nicholson, W.L., Munakata, N., Horneck, G., Melosh, H.J. and Setlow, P. (2000) Resistance of *Bacillus* endospores to extreme terrestrial and extraterrestrial environments. *Microbiology and Molecular Biology Reviews*, **64**, 548–572.

Nikiyan, H., Vasilchenko, A. and Deryabin, D. (2010) Humidity-dependent bacterial cells functional morphometry investigations using atomic force microscope. *International Journal of Microbiology*, Article ID 704170, 1–5.

Norton, T. and Sun, D.-W. (2008) Recent advances in the use of high pressure as an effective processing technique in the food industry. *Food and Bioprocess Technology*, **1**, 2–34.

Oey, I., Lille, M., van Loey, A. and Hendrickx, M. (2008a) Effect of high-pressure processing on colour, texture and flavour of fruit- and vegetable-based food products: a review. *Trends in Food Science and Technology*, **19**, 320–328.

Oey, I., van Der Plancken, I., van Loey, A. and Hendrickx, M. (2008b) Does high pressure processing influence nutritional aspects of plant based food systems? *Trends in Food Science and Technology*, **19**, 300–308.

Oliveira, A.C., Gaspar, L.P., Da Poian, A.T. and Silva, J.L. (1994) Arc repressor will not denature under pressure in the absence of water. *Journal of Molecular Biology*, **240**, 184–187.

Olivier, S.A., Bull, M.K. and Chapman, B. (2012) *Bacillus* spp. spores produced at lower temperatures are more resistant to high pressure thermal processes but mineralization does not predict relative resistance. *Innovative Food Science and Emerging Technologies*, **16**, 96–101.

Pehl, M., Werner, F. and Delgado, A. (2002) Experimental investigation on thermofluid dynamical processes in pressurized substances. *Progress in Biotechnology*, **19**, 429–435.

Perrier-Cornet, J.M., Hayert, M. and Gervais, P. (1999) Yeast cell mortality related to a high-pressure shift: occurrence of cell membrane permeabilization. *Journal of Applied Microbiology*, **87**, 1–7.

Perrier-Cornet, J.M., Marechal, P.A. and Gervais, P. (1995) A new design intended to relate high pressure treatment to yeast cell mass transfer. *Journal of Biotechnology*, **41**, 49–58.

Pope, D.H., Smith, W.P., Swartz, R.W. and Landau, J.V. (1975) Role of bacterial ribosomes in barotolerance. *Journal of Bacteriology*, **121**, 664–669.

Rasanayagam, V., Balasubramaniam, V.M., Ting, E., Sizer, C.E., Bush, C. and Anderson, C. (2003) Compression heating of selected fatty food materials during high-pressure processing. *Journal of Food Science*, **68**, 254–259.

Rastogi, N.K., Angersbach, A. and Knorr, D. (2000) Synergistic effect of high hydrostatic pressure pretreatment and osmotic stress on mass transfer during osmotic dehydration. *Journal of Food Engineering*, **45**, 25–31.

Rastogi, N.K. and Niranjan, K. (1998) Enhanced mass transfer during osmotic dehydration of high pressure treated pineapple. *Journal of Food Science*, **63**, 508–511.

Rastogi, N.K., Raghavarao, K.S., Balasubramaniam, V.M., Niranjan, K. and Knorr, D. (2007) Opportunities and challenges in high pressure processing of foods. *Critical Reviews in Food Science and Nutrition*, **47**, 69–112.

Rawson, A., Patras, A., Tiwari, B.K., Noci, F., Koutchma, T. and Brunton, N. (2011) Effect of thermal and non thermal processing technologies on the bioactive content of exotic fruits and their products: review of recent advances. *Food Research International*, **44**, 1875–1887.

Reineke, K., Mathys, A. and Knorr, D. (2011) The impact of high pressure and temperature on bacterial spores: inactivation mechanisms of *Bacillus subtilis* above 500 MPa. *Journal of Food Science*, **76**, M189–M197.

Rendueles, E., Omer, M.K., Alvseike, O., Alonso-Calleja, C., Capita, R. and Prieto, M. (2011) Microbiological food safety assessment of high hydrostatic pressure processing: a review. *LWT - Food Science and Technology*, **44**, 1251–1260.

Sale, A.J.H., Gould, G.W. and Hamilton, W.A. (1970) Inactivation of bacterial spores by hydrostatic pressure. *Journal of General Microbiology*, **60**, 323–334.

Sharma, A. (2011) Microbial adaptation to high pressures – from denial to a new paradigm shift. *AGU Fall Meeting Abstracts*, **1**, 04.

Sun, D.-W. (2005) *Emerging Technologies for Food Processing*. Elsevier Academic Press, Amsterdam, Boston.

Tauscher, B. (1995) Pasteurization of food by hydrostatic high pressure: chemical aspects. *Zeitschrift für Lebensmittel-Untersuchung und Forschung*, **200**, 3–13.

Thakkar, S.D. (2012) Effect of processing on the antioxidant activity and other quality parameters of Muscadine grape juice. MS Thesis, Rutgers University, New Brunswick, New Jersey, USA.

Thwaites, J.J. and Mendelson, N.H. (1985) Biomechanics of bacterial walls: studies of bacterial thread made from *Bacillus subtilis*. *Proceedings of the National Academy of Science USA*, **82**, 2163–2167.

Thwaites, J.J. and Surana, U.C. (1991) Mechanical properties of *Bacillus subtilis* cell walls: effects of removing residual culture medium. *Journal of Bacteriology*, **173**, 197–203.

Wong, P.T.T. and Heremans, K. (1988) Pressure effects on protein secondary structure and hydrogen deuterium exchange in chymotrypsinogen: a Fourier transform infrared spectroscopic study. *Biochimica et Biophysica Acta (BBA) – Protein Structure and Molecular Enzymology*, **956**, 1–9.

Yen, G.-C. and Lin, H.-T. (1996) Comparison of high pressure treatment and thermal pasteurization effects on the quality and shelf life of guava puree. *International Journal of Food Science and Technology*, **31**, 205–213.

25
Ozone Processing

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25.1 Introduction

Ozone has been recognized as a promising food processing agent, which is gaining interest in the food industry due to its oxidant properties. Ozone has been successfully applied in diverse fields such as food decontamination, disinfection of plant equipment and waste water treatment. This fact, together with consumers' rising demand for minimally processed products and the frequent outbreaks of food-related illnesses, has led to several researches aiming at understanding the commercial exploitation of this technology. With the efficacy and usefulness of ozone proved over the years in water treatment, in surface cleaning and disinfection, and in food decontamination, the regulatory status of ozone for food processing applications have to be developed in line with process validation.

Although the potential of ozone to disinfect polluted water was recognized in Europe in 1886, only in 1982 did the US Food and Drug Administration (FDA) grant GRAS (generally recognized as safe) status for ozone as a disinfectant of bottled water (O'Donnell *et al.*, 2012). In 1997, an Expert Panel of Food Scientists convened by the Electric Power Research Institute (EPRI) supported a GRAS classification of ozone as a disinfectant for foods when used at levels and by methods of application in accordance with good manufacturing practices (GMPs). Even though the FDA did not object this GRAS status, in 2000 a Food Additive Petition filled by the EPRI requested FDA approval of ozone for direct contact with foods. Only in 2001 did the FDA finally approve the application of ozone as a direct food additive for the treatment, storage and processing of foods in gas and aqueous phases (FDA, 2001).

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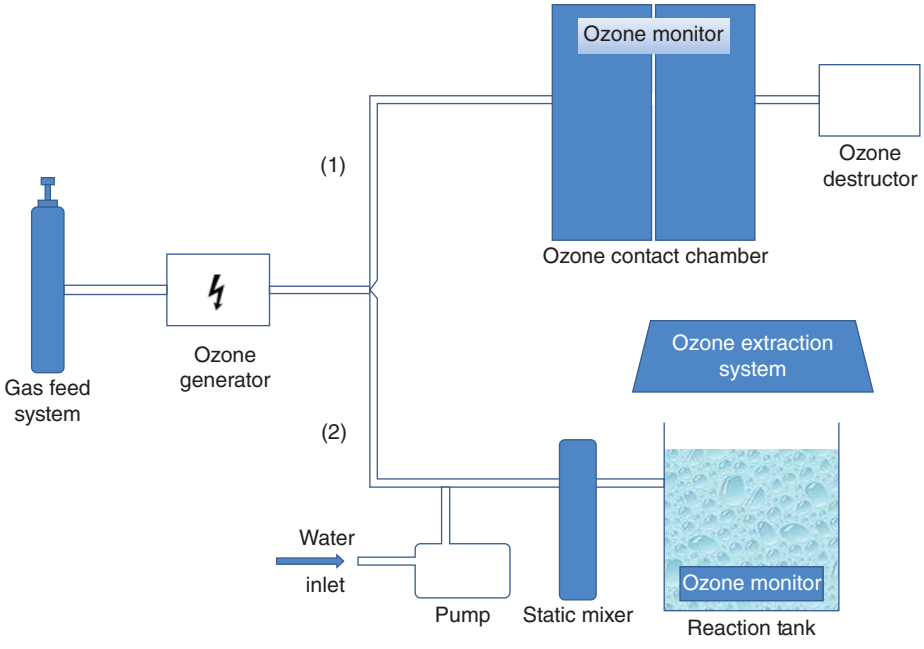

Figure 25.1 Simplified ozone system schematic: (1) gaseous and (2) aqueous applications

Since then, several studies have been conducted to evaluate the effectiveness of ozone in several food products. The ozone treatments are mainly applied in two forms: (1) gaseous ozone can be added continuously or intermittently to an atmosphere where the product is stored or (2) the product can be washed or dipped in water containing ozone (Figure 25.1). However, when the food is a fluid product, such as juices, the addition of gaseous ozone is made directly into the fluid. Indeed, this kind of product has been associated with several outbreaks of illness, which warns that the microbiological control of the processes cannot be neglected. The juice processing requirements are regulated by the FDA, which imposes the application of treatments capable of consistently achieving at least a 5-log reduction in the level of the most resistant pathogen in the specific juice (FDA, 2004).

The antimicrobial effect of ozone is of the utmost importance in the control of pathogenic and spoilage microorganisms. Despite the fact that its efficacy has been demonstrated for many contaminated food products, dissimilar results are often reported (discussed latter). Concerning quality aspects, it is desirable that there should be a high retention of physicochemical, sensorial and nutritional attributes of the products treated with ozone (the impacts of ozone on food quality characteristics are presented later). Only a conjoint control of ozone action on safety and quality features can provide safe products

from a microbiological perspective, with an extended shelf-life meeting the required quality standards.

Applications of ozone in the food industry are available (O'Donnell *et al.*, 2012; Chiattone, Torres and Zambiazi, 2008; Guzel-Seydim, Bever and Greene, 2004; Guzel-Seydim, Greene and Seydim, 2004); in more detail these are given for fruits and/or vegetables (Miller, Silva and Brandão, 2013; Cullen *et al.*, 2010; Karaca, 2010; Karaca and Velioglu, 2007) and seafood (Blogoslawski and Stewart, 2011; Goncalves, 2009). Nevertheless, ozone applications are also effective in other products, such as meat, poultry, dry foods, cereals and dairy products. The challenge is to improve the acceptance of this technology by the food industry, through dissemination of the results obtained from academic research, awareness of related process costs and environmental impact.

25.2 Ozone properties

Ozone is a highly unstable triatomic oxygen molecule (O_3), arranged to form an obtuse angle, with a molecular weight of 47.98 g/mol. At a normal pressure of 1.013 bar, liquid O_3 density is 1352 kg/m^3 with boiling and melting points of −111.3 and −192.5 °C, respectively, whereas in the gaseous phase, O_3 has a density of 2.141 kg/m^3 at 1.013 bar pressure and 0 °C temperature (Cullen and Tiwari, 2012).

Although ozone is relatively stable in the gaseous state, it is highly unstable in aqueous solution. At room temperature of 20 °C, the typical ozone half-life in the gaseous state is around 3 days, which is longer than the ozone half-life in dissolved water (≈ 20 minutes) for the same temperature (Goncalves, 2009). In water, ozone quickly degrades to oxygen. Although ozone is extremely soluble in water (ozone solubility is 580 mg/L at 27 °C), its solubility rate is dependent on several factors, such as pressure, temperature, pH, ozone bubble size, flow rate of ozone and contact time, purity of water and also on the technology of interchange of gas/liquid (Khadre, Yousef and Kim, 2001).

At room and cold temperatures, ozone is a gas with a pungent characteristic odour, which is detectable by humans at concentrations as low as 0.02 ppm (Horváth, Bilitzky and Huttner, 1985). With an oxidation-reduction potential of 2.07 V, ozone is eligible as one of the strongest and most reactive known sanitizers. This value makes ozone 1.5 times stronger than chlorine against several microorganisms. However, chlorine and associated compounds remain to be the most commonly used sanitizers, even when their use results in the formation of various chlorinated by-products, some of which are considered carcinogenic (Wei *et al.*, 2007; Kim, Yousef and Chism, 1999). Ozone is considered an alternative over chlorine due to several advantages: (1) ozone can be generated on-site; (2) it spontaneously and rapidly decomposes into nontoxic products such as oxygen and water, leaving no traces; (3) it does not produce toxic

halogenated compounds; (4) its action is very rapid, with a good penetration ability; (5) it is effective at short contact times; and (6) it is effective against a wide range of microorganisms. This last item has been a point of concern for the users of chlorine, since some of the concerned foodborne pathogens are found to be resistant to chlorination (Parish *et al.*, 2003).

Ozone has also the significant advantage of being an environmental friendly technology. The potential of ozone for wastewater treatment and water reuse in the food industry reduces the company's environmental costs and at the same time facilitates their compliance with statutory obligations (Patil and Bourke, 2012).

Goncalves (2009) gathered information about how ozone acts as an oxidant and stated that three major action pathways can occur: (1) direct oxidation reactions of ozone, resulting from the action of an oxygen atom; these reactions are typically first order with high redox potential; (2) indirect oxidation reactions of ozone, where the ozone molecule decomposes to form free radicals, which will quickly react to oxidize organic and inorganic compounds; and (3) ozone may also act by ozonolisys, by fixing the complete molecule on double-linked atoms, producing two simple molecules with different properties and molecular characteristics.

25.3 Ozone generation

Ozone is formed by a high-energy input that splits the molecular oxygen (O_2) into two free oxygen radicals (O^-). These free oxygen radical molecules rapidly react with available oxygen to form O_3. The source of this high energy is usually ultraviolet radiation or electrical (corona) discharge. In order to generate commercial levels of ozone, the corona discharge method is generally used (Guzel-Seydim, Greene and Seydim, 2004). However, there are other less mainstream methods of ozone generation, such as electrochemical and radiochemical ones. However, they have limited use due to the very high associated costs and/or to the low ozone yield achieved.

25.3.1 Corona discharge method

Corona discharge is a method that consumes high energy. It is, however, the most commonly used one, because it allows generation of commercial ozone levels. In this method, the feed gas (such as dry air, oxygen or a gaseous mixture) is passed through an electrical field. This electrical field consists of two high-voltage electrodes separated by a dielectric material (that controls and maintains the electrical discharge), which is usually ceramic or glass. When a voltage is supplied to the electrodes, a corona discharge forms between the two electrodes and the feed gas in the discharge gap becomes partially ionized, being converted to ozone. Indeed, this corona discharge ruptures the

stable oxygen molecule and forms two oxygen radicals that can combine with oxygen molecules to form ozone. About 85% of the electrical energy supplied to a corona discharge ozone generator produces heat that must be removed. This excessive heat is often removed by cooling water or air.

The yield of ozone generation is directly dependent on several factors, such as feed gas concentration, power input and cooling water temperature. To attain maximum yield and to minimize the energy involved, it is important to be aware of the following:

1. Ozone is produced from oxygen, so it can be produced from ambient air (21% oxygen) or nearly pure oxygen (e.g. 95%). The amount of ozone produced is dependent on the oxygen concentration in the gas feeding the corona. If air is passed through the generator as a feed gas, 1–3% of ozone can be produced. When pure oxygen is used as the feed gas, ozone production may reach yields up to 6% (Rice, Farquhar and Bollyky, 1982).
2. The amount of energy applied to the gas and the gap between the electrodes are critical to the concentration of ozone produced. It is a combination of the voltage and frequency that results in a given energy input. Typically, voltages between 7 and 30 kV are used with frequencies ranging from the mains supply of 50 or 60 Hz, medium up to 1000 Hz and high up to 4000 Hz (Vaduganathan, Poonamallie and Nagalingam, 2012).
3. As already mentioned, ozone generation is an inefficient process whereby a significant part of the applied energy is wasted as heat. It is of main importance to remove this heat by proper cooling, since ozone is destroyed at high temperatures. To limit the decomposition of ozone and at the same time allow its formation, the temperature in the discharge gap must be maintained at a moderate range (Muthukumarappan, 2012).

25.3.2 Ultraviolet method

In the ultraviolet (UV) method, ozone is formed throughout oxygen exposure to UV radiation at a wavelength of 140–190 nm. When an oxygen molecule (O_2) is passed over a UV light, it will absorb the light energy and dissociates (O^-), being ready to combine with other oxygen molecule (O_2) to form ozone. Although a wide range of UV bulbs are available, the wavelength range varies from 180 to 254 nm. Nevertheless, each wavelength of light favours different reactions and their quantum yield. The breakdown of the oxygen molecule has a higher yield at wavelengths less than 200 nm. However, like oxygen, ozone also absorbs light. The dissociation or photolysis of ozone has its greatest yield between 200 and 308 nm. Therefore, for an effective ozone production, it is necessary to utilize a short wavelength near 185 nm. In practice, since these lamps can only produce ozone up to 0.3–0.4% by weight (Cullen and Tiwari, 2012), the UV method can only be feasible where production of small amounts of ozone is desired (e.g. laboratories and pilot plants).

25.3.3 Electrochemical (cold plasma) method

The electrochemical method usually applies an electrical current, between an anode and cathode, in an electrolytic solution containing water and a solution of highly electronegative anions. This results in the production of a mixture of oxygen and ozone at the anode (Mahapatra, Muthukumarappan and Julson, 2005). Work has been done with different electrolytes and anode materials to improve the efficiency of production and minimize the corrosive reactions on the anode surface. Some of the advantages of this method are: (1) the use of low-voltage direct current; (2) no feed gas preparation; (3) reduced equipment size; (4) high concentration of ozone (at least 10%) can be achieved; and (5) generation in water (O'Donnell *et al.*, 2012). This ozone generation method has been making several improvements that will make it commercially viable in a near future, especially for potable or wastewater treatment.

25.3.4 Radiochemical method

In the radiochemical method, ozone is formed through the oxygen dissociation by high-energy irradiation. This energy is usually accomplished by radioactive rays of the isotopes ^{137}Cs, ^{60}Co or ^{90}Sr (Heim and Glas, 2011). This method has a favourable thermodynamic yield, since 35% of the available energy is used for the decomposition of oxygen molecules. However, due to process drawbacks and the inherent danger of radioactive contamination, in practice this procedure is rarely used.

25.4 Antimicrobial action

There are a wide variety of microorganisms commonly associated with food contamination, including bacteria (some pathogenic), viruses, parasites and fungi/moulds. Each food product has its native microflora and is susceptive to contamination with different kinds of microorganisms. Therefore, it is of great importance for the assessment of ozone efficacy for the elimination of target microorganisms.

Since ozone is highly unstable, it does not maintain a high residual level, being generally used as a primary disinfectant. This often requires the use of other chemical agents or techniques in combination with ozone to attain sufficient levels of disinfection in food products (Kim, Yousef and Dave, 1999). Nevertheless, the effectiveness of ozone treatment on microbial inactivation depends on several factors. The main influential factors are: type of product, target microorganism, initial microbial load level and physiological state of the bacterial cells, and the ozone physical state. The sensitivity of microorganisms to ozone is also extremely affected by environmental factors such as medium

pH, temperature, humidity, the presence of additives (e.g. acids, surfactants and sugars) and the amount of organic matter.

25.4.1 Inactivation mechanisms

Although research on ozone inactivation mechanisms against many microorganisms has been carried out, the microbicide reaction of ozone is not yet clearly known, due to the complexity of processes involved.

Molecular ozone or its decomposition products progressively oxidize vital cellular components of microorganisms including proteins, unsaturated lipids and respiratory enzymes in cell membranes, peptidoglycans in cell envelopes, enzymes and nucleic acids in the cytoplasm and proteins and peptidoglycan in spore coats and virus capsids (Khadre, Yousef and Kim, 2001). It has been suggested that cell surface is the primary target of ozonation, via oxidation of amino acids of bacterial cell membrane, by oxidation of cell wall glycoproteins and glycolipids or by the degradation of unsaturated lipids of the cell envelope (Thanomsub *et al.*, 2002). As a result, a large part of the membrane barrier is destroyed, with the subsequent cell disruption and leakage of cellular contents, followed by bacterial cells lyses. When this is not enough for the immediate cell destruction, ozone can penetrate inside the bacterium and oxidize other essential components, for example enzymes, proteins and nucleic acids. This was confirmed by Komanapalli and Lau (1996), who reported that the membrane permeability of *Escherichia coli* cells was compromised by short-term ozone exposure (1–5 min), affecting both lipid and protein components. However, cell viability was unaffected and the intracellular components, proteins and DNA remained intact. With longer ozone exposures (up to 30 min), cells viability decreased, with a progressive degradation of intracellular proteins and DNA.

25.4.2 Inhibitory spectrum

Bacteria It has been proven that ozone is effective in inactivating several bacteria, which include Gram-negative and Gram-positive, vegetative cells and spore forms. However, the inherent sensitivity of the bacteria to ozone depends on the microorganism species and on its physiological state. For instance, Gram-positive bacteria are generally more resistant to ozone than Gram-negative ones. The explanation can be the difference in cell wall structure of the two types of bacteria. Cell walls of Gram-positive bacteria consist of many layers of peptidoglycans, forming a thick rigid structure, whereas cells walls of Gram-negative bacteria consist of an outer membrane, which contains lipoprotein and lipopolysaccharides and only a few layers of peptidoglycans beneath (Thanomsub *et al.*, 2002). As already mentioned,

the lipoprotein and lipopolysaccharide layers are the prime sites of destruction, resulting in increased cell permeability and eventual cell lysis of the Gram-negative bacteria (Kim, Yousef and Dave, 1999). The higher resistance of Gram-positive bacteria can be partly explained by the greater amount of peptidoglycan in their cell walls. This is supported by the study of Rey *et al.* (1995), who concluded that *N*-acetyl glucosamine, a compound present in peptidoglycan of bacterial cell walls, was resistant to the action of ozone in aqueous solutions with pH between 3 and 7.

Ozone is also known to be more effective against vegetative bacterial cells than against bacterial spores. Bacterial spores are known to be extremely resistant to environmental abuses and toxic chemicals. When compared to vegetative cells, bacterial spores showed greater resistance to ozone treatments (Mahfoudh *et al.*, 2010; Akbas and Ozdemir, 2008). Although many factors may explain this resistance, the major contribution appears to be the multilayered spore coat (Young and Setlow, 2004). Spores of *Bacillus* species are the most studied ones, due to their specific and proven resistance to ozone (Ijabadeniyi, Minnaar and Buys, 2011).

Independently of the structural differences of many bacteria, it is still not clear whether the ozone effects are due to the reactions of molecular ozone itself or to the products that are produced by its decay to molecular oxygen. Among the products produced, the hydroxyl radical is one of the strongest known oxidizers (Perry and Yousef, 2011). Although Cho, Chung and Yoon (2002) found that these hydroxyl radicals were the main factors responsible for the inactivation of *Bacillus subtilis* spores, no difference in *E. coli* inactivation was observed by Hunt and Marinas (1997) when cells were treated with ozone in the presence and absence of radical scavenging compounds. This finding was indicative of the fact that dissolved molecular ozone was primarily responsible for *E. coli* inactivation in the range of experimental conditions investigated.

Several researchers compared the efficiency of gaseous ozone with aqueous ozone on bacterial inactivation. Contradictory results have been reported, since some researchers verified that water containing ozone was significantly more effective (Zorlugenc *et al.*, 2008; Kim, Yousef and Chism, 1999) and others obtained higher microbial log reductions when gaseous ozone treatment was applied (Singh *et al.*, 2002). These results showed the importance of considering all process conditions in the assessment of ozone effectiveness for a given food product and target microorganism. Some of these issues will be discussed in Section 25.4.3.

Fungi and their mycotoxins This group is a focus of concern, not only because they cause product deterioration but also due to their potential for mycotoxin production. Post-harvest decay is mainly associated with fungi such as *Aspergillus* and *Penicillium*. These fungi, together with *Fusarium*, are the main mycotoxin producers, especially aflatoxins, ochratoxin A and

patulin, frequently found in cereals, pulses, nuts and their products. Several studies support the use of ozone for the inactivation of these mycotoxigenic fungi and in the degradation of their mycotoxins. Those works were reviewed by Freitas-Silva and Venancio (2010) and, based on their findings, the researchers suggested that the mechanism involved in fungi inactivation by ozone is also related, with implications in membrane integrity.

In general, ozone is referred to as an effective fungicide with a germicidal effect, usually greater for moulds than for bacteria. However, as happens to bacteria, the effectiveness of ozone may vary among fungi species. This was confirmed by Palou *et al.* (2001), who observed that although the growth of *Penicillium italicum* was inhibited by ozone, *Penicillium digitatum* was ozone resistant.

Most of the applications of ozone for fungi inactivation and mycotoxins degradation use ozone in the gaseous state. Zorlugenc *et al.* (2008) compared the effectiveness of gaseous and aqueous ozone on fungi inactivation and on the removal of aflotoxin B1 from dried figs. Results showed that gaseous ozone was more effective than ozonated water on aflatoxin B1 reduction, whereas aqueous ozone was found to be more effective in fungi inactivation.

Viruses Ozone is potentially an effective virucidal agent, since relatively low ozone concentration and short contact time are enough to inactivate viruses (Kowalski, Bahnfleth and Whittam, 1998). Although the number of studies on virus inactivation with ozone is limited, it has been demonstrated that ozone can destroy a wide range of viruses including hepatitis A and influenza A (Hall and Sobsey, 1993; Herbold, Flehmig and Botzenhart, 1989). While there are some discrepancies as to whether virus sensitivity to ozone is comparable to bacteria, the majority of researchers affirmed that the bacteriophages are the least resistant to ozone, followed by polioviruses, whereas human rotavirus is the most resistant one (Khadre, Yousef and Kim, 2001; Hall and Sobsey, 1993). It has been reported that ozone interacts and modifies proteins in the virus capsid, which are used by the virus to attach to the cell surface. Kim, Gentile and Sproul (1980) studied the inactivation of bacteriophage f2 upon reaction with ozone and suggested that ozone attacks the capsid protein, which resulted in the release of ribonucleic acid (RNA), and consequently disrupted adsorption to the host pili. The RNA, not protected by the phage coat, is further susceptible to oxidation by ozone. A similar mode of action was proposed for tobacco mosaic virus (TMV), where ozone attacked both the capsid and RNA. Damaged RNA forms cross-links with the capsid subunits to cause a loss of virus activity (Shinriki *et al.*, 1988).

25.4.3 Influential factors on ozone antimicrobial action

As already mentioned, the microbial sensitivity to ozone, and hence its effectiveness for inactivation, is dependent not only on the microorganism but also

on other process conditions, such as ozone concentration, contact time, temperature and pH of the medium, the ozone physical state and application method, the presence of organic matter and the type and characteristics of the food product to treat. In relation to ozone concentration, it is important that not only the correct amount of ozone is applied but also that the residual ozone is present in the medium. Residual ozone is the concentration of ozone that can be detected in the medium after application to the target surface. Both the instability of ozone under certain conditions and the presence of ozone-consuming materials affect the level of residual ozone available in the medium (Pascual, Llorca and Canut, 2007). Aspects such as the accuracy of ozone quantification methods and the assessment of ozone antimicrobial effectiveness are important for industrial applications of this sanitizer.

Temperature It is known that as temperature increases, the solubility and stability of ozone decreases and the ozone decomposition rate increases. However, there is no consensus about the effect of temperature on the biocidal efficacy of ozone. Patil *et al.* (2009b) studied four different temperatures (12–15, 20, 25 and 30 °C) on *E. coli* inactivation by ozone, concluding that antimicrobial activity of ozone decreases with an increase in the temperature. Achen and Yousef (2001) used ozone at 4, 22 and 45 °C in *E. coli* contaminated apples and observed that counts of the bacterium on the fruit surface decreased 3.3, 3.7 and 3.4 log units, respectively. However, statistical analysis showed no significant differences between the three results. The residual ozone concentration was higher at the lowest temperature and decreased with temperature increase. The authors suggested that when temperature is increased, the increase in ozone reactivity compensated for the decrease in its stability, and thus no appreciable change in ozone efficacy was detected. In contrast, Kim (1998) noted that ozone was more effective at 24 °C than at 7 °C. These results support the fact that the simultaneous contribution of the two factors (solubility/stability and reactivity) can vary with experimental conditions, making it difficult to predict the influence of temperature on a particular application (Pascual, Llorca and Canut, 2007).

pH Ozone is more stable at low pH than at high pH values. The effect of pH on ozone microbial inactivation is mainly attributed to the fact that the ozone decomposition rate changes substantially with pH variations. When the pH is low, microbial inactivation is mostly through reaction with molecular ozone. At a high pH, the faster ozone decomposition results in the formation of several types of oxidants with different reactivities. The different reaction rates of these radical species are responsible for the changes of the microbicidal action of ozone.

Humidity Several studies reported that at a low relative humidity ozone showed no germicidal potential. Kim, Yousef and Dave (1999) found that

200 ppm ozone and water activity of 0.85 produced no effect on the microbial load, while up to 10^5 CFU/g reductions were achieved when water activity was 0.95. This effect was observed for bacteria, fungi and spores.

Food product Ozone reacts with other compounds. Food systems are rich in organic matter, which will compete with microorganisms for ozone demand, and thus ozone disinfection efficiency is reduced. Nevertheless, Restaino *et al.* (1995) concluded that the type of organic material present in an ozonation process is more important than the amount of organic matter. Guzel-Seydim, Bever and Greene (2004) evaluated the efficacy of ozone (at 0.4 ppm for 10 min) to reduce *Bacillus stearothermophilus* spores, vegetative cells of *E. coli* and *Staphylococcus aureus* in the presence of fat, protein and carbohydrate sources. They concluded that food components had a significant effect on the bacteriocidal power of ozone against the microorganisms. Starch provided little or no protective effects compared to the buffer control and locust bean gum provided an intermediate level of protection, while caseinate and whipping cream provided the greatest levels of protection to the bacterial populations. Patil *et al.* (2009a) also reported that the inactivation rate of *E. coli* was strongly dependent on the soluble solids content present in orange juice. They suggested that sugars, fibre and ascorbic acid could act as protective barriers for microbial cells against ozone. This fact is related to ozone accessibility to the targeted microorganisms. It is known that bacteria can be associated with suspended particles or cellular components, which may reduce the effectiveness of the ozone treatment applied to a food product. Although some studies reported that the antimicrobial effect of ozone was enhanced upon application of sonication to break down lumps of microorganisms, such an effect was not observed during the ozone treatment of fresh lettuce (Kim, Yousef and Dave, 1999). It was proposed that sonication might enhance the decomposition of ozone itself or increase ozone demand by detaching organic materials from the cut surfaces of the shredded lettuce.

The type of food product and surface characteristics also influence the effectiveness of an ozone treatment. Long *et al.* (2011) concluded that the folding and indentations on tomato and green onion surfaces allowed bacteria attachment that enabled their survival after decontamination treatments.

25.5 Applications of ozone

Although ozone has been used effectively in water treatments, its use in food processing has only recently come to the fore. This was essentially the result of FDA approval of ozone. Currently, ozone is used in food processing industries all over the world as a food antimicrobial agent, and as a cleaning and disinfection agent for equipment and facilities (Figure 25.2).

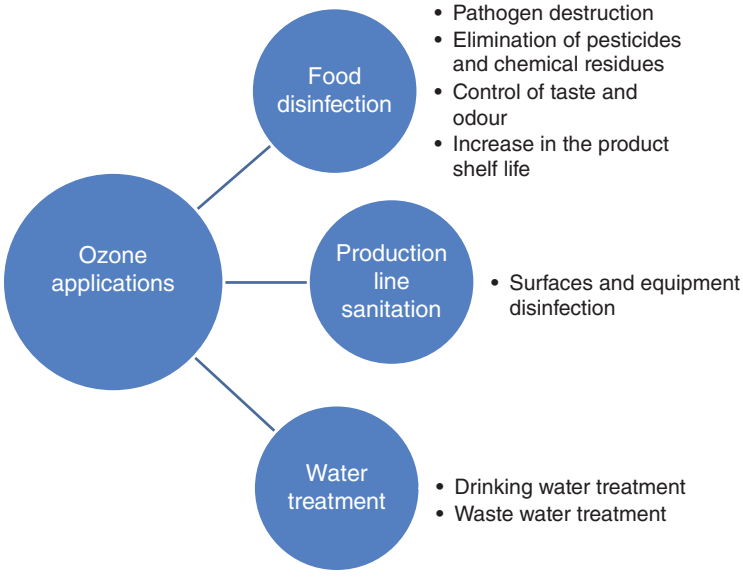

Figure 25.2 Possible ozone applications in the food industry

25.5.1 Impact on microbiological and quality characteristics of some products

Ozone acts as a food decontaminant, and the impact of ozone on quality features of the products is important. Ozone process conditions should simultaneously guarantee microbial elimination and prevent, as far as possible, significant losses of quality attributes. The impact of ozone on microbiological contamination and on key quality characteristics of water and foods will be discussed in subsequent sections.

Water Ozone can be applied in potable water, wastewater treatments and for water reuse in the food industries. The major purposes of its utilization are:

1. Disinfection. Ozone is a powerful oxidizing agent, which is even toxic at low concentrations and low contact time for most waterborne organisms. Possible hazard pathogens present in water include viruses, bacteria such as *Salmonella*, *Cholera*, *Campylobacter* and *Shigella*, and protozoa like *Giardia lamblia* and other *Cryptosporidia*. However, ozone can only be used as a primary water disinfectant, since it has no residual effects in the distribution system due to its short half-life. For this reason, ozone should be coupled with other secondary disinfectants such as chlorine or chloramines for a complete disinfection system (Rakness, 2005).

2. Iron and manganese removal. Iron and manganese, when presented in concentrations above certain limits (0.1 mg/L for iron and 0.05 mg/L for manganese), are associated with unpleasant flavour and colour characteristics of water. Ozone can oxidize the soluble inorganic compounds (Fe^{2+} and Mn^{2+}) to Fe^{3+} and Mn^{4+}, which can be hydrolysed and precipitate as $Fe(OH)_3$ and MnO_2. The reactions consume 0.43 and 0.88 mg of ozone per mg of Fe^{2+} and Mn^{2+}, respectively (Langlais, Reckhow and Brink, 1991).
3. Control of taste and odour. Off-taste and odour are related to the presence of organic compounds or synthetic organic compounds, such as solvents and pesticides, even at low concentrations. The organic compounds may result from the decay of plant matter or can be produced by the activity of actinomycetes and blue-green algae (Langlais, Reckhow and Brink, 1991). The major by-products of algae are 2-metil-isoborneol (MIB) and geosmin, which are organoleptically detectable in very low concentrations (of the magnitude of nanograms per litre). Ozone reduces and/or eliminates these undesirable tastes and odours by oxidation of the compounds.
4. Control of colour. Surface waters have natural organic materials such as humic and fulvic acids and tannin that give coloration. These compounds result from the decomposition of vegetable matter and are molecules containing many conjugated double bonds, some of which are easily broken by ozone.
5. Particulate and turbidity removal. Water turbidity is removed by ozonation through a combination between chemical oxidation and charge neutralization. The colloidal particles that cause turbidity are kept in suspension due to negative charges. When ozone is applied, the charge is neutralized and the colloid is broken down. Additionally, ozone alters the colloids surface properties because it oxidizes the organic materials present on it.

As already mentioned, many organic compounds present in water are oxidized by ozone. The by-products of oxidation are usually more easily biodegraded. However, if they are not removed during water treatment this can lead to biological regrowth problems. One of the most important by-products of ozonation is bromate, which is formed in bromide-containing water. This compound has been identified as potentially carcinogenic and therefore its maximum concentration level has been regulated as 10 μg/L (Rakness, 2005).

Fruits and vegetables Fruit and vegetable contamination is mostly associated with bacteria such as *Salmonella*, *E. coli*, *Listeria monocytogenes*, *Shigella*, *Campylobacter*, *Clostridium botulinum* and *Bacillus cereus,* some viruses like Norwalk and hepatitis A, and also some fungi such as *Aspergillus* and *Penicillium*. Without any efficient decontamination treatment applied before

consumption, fruits and vegetables can cause serious health issues and their shelf-life is reduced due to the perishable nature of these products. For this reason, antimicrobial efficacy of ozone has been studied for a number of produce commodities against several microorganisms. The effect of ozone (in its gaseous and aqueous forms) on the quality of these products has been also evaluated by many researchers.

Ozonated water has been used to wash fruits and vegetables surfaces, aiming at decontamination. In apples, the procedure was effective in the inactivation of *E. coli* and *L. monocytogenes* (Rodgers *et al.*, 2004; Klingman and Christy, 2000). Achen and Yousef (2001) compared a method of bubbling ozone in water during apple washes by dipping apples in pre-ozonated water. They concluded that bubbling ozone in water was more effective in reducing microbial contamination.

Another fruit that has been extensively studied is strawberry, due to its short shelf-life. Demirkol *et al.* (2008) reported that the optimal conditions for strawberry disinfection were either exposure to 40 ppm of gaseous ozone or dipping in ozonated water at 8 ppm for 30 min. Strawberries washed in ozonated water (20 ppm) for 3 min and stored for 12 days at 4 °C produced reductions in mesophiles, psychotrophes and yeasts/moulds of 1.28, 1.51 and 0.78 log CFU/g, respectively (Wei *et al.*, 2007). Changes in colour and firmness were not detected after ozone treatment and throughout storage. Bialka and Demirci (2007) studied the effect of ozone concentration in water and of temperature and contact time on *E. coli* and *Salmonella* loads for strawberries and raspberries. The conditions were ozone concentrations of 1.7–8.9 ppm at 20 °C for 2 to 64 min or 21 ppm at 4 °C for 64 min. For raspberries, maximum reductions were attained at 4 °C (5.6 and 4.5 log CFU/g for *E. coli* and *Salmonella*, respectively). For strawberries, the higher load reductions were observed at 20 °C after 64 min (2.9 and 3.3 log CFU/g for *E. coli* and *Salmonella*, respectively). These results reinforce the condition that ozone effectiveness must be assessed for each combination of product/microorganism of concern.

Disinfection of tomatoes using ozonated water has also been much investigated. Venta *et al.* (2010) suggested that washing tomatoes in ozonated water (1 ppm) for 15 min is an adequate disinfection procedure. Lower ozone concentrations (0.05–0.2 ppm) were tested by Long *et al.* (2011), who verified that these conditions were effective in reducing aerobic bacteria contamination, but did not significantly reduce *E. coli* or *Salmonella*.

Concerning the effect of aqueous ozone on microbial contamination, lettuce is probably the mostly studied food product (Olmez and Akbas, 2009; Akbas and Olmez, 2007; Wei *et al.*, 2007). To be effective, ozone should be applied at high concentrations, combined with other disinfectants (Yuk *et al.*, 2006; Garcia, Mount and Davidson, 2003; Singh *et al.*, 2002) or with high-speed stir (Kim, Yousef and Chism, 1999). In general, ozone treatments result in good retention of products' sensorial quality (colour, texture, visual

appearance, aroma, off-flavour and off-odour) and browning control with no detrimental reduction of antioxidant constituents (Olmez and Akbas, 2009; Akbas and Olmez, 2007; Baur *et al.*, 2004).

Gaseous ozone is often used as a protective storage atmosphere against mould and bacteria. The fruits most susceptible to this kind of contamination are grapes and strawberries. Tzortzakis, Singleton and Barnes (2007) assessed the impact of an ozone-enriched atmosphere (0.1 μmol/mol) on tomatoes, strawberries, grapes, plums and clementines inoculated with *Botrytis cinerea* (gray mould) and stored for 13 days (at 13 °C and 95% relative humidity). Although the results suggested that the reaction to, and benefits of, ozone treatment varied with commodities, fungal development was markedly suppressed in all infected fruits exposed to low levels of ozone, even for short exposure periods.

Many fruits release ethylene gas, which speeds up the ripening process. Ozone removes ethylene from air, thereby extending the shelf-life of many ethylene-sensitive produce during storage. Minas *et al.* (2012) observed that cold storage (at 0 °C under 95% relative humidity) of kiwifruit with a gaseous ozone atmosphere (0.3 ppm) for up to 5 months improved the post-harvest behaviour of the fruit. Ozone blocked the ethylene production, delayed ripening and stimulated antioxidant and antiradical activities of kiwifruit. Beyond these positive effects, ozone also promoted higher firmness retention.

Many studies using gaseous ozone were conducted in cantaloupe melons and spinach due to recent outbreaks of *Salmonella* and *E. coli*, respectively. Selma *et al.* (2008a) submitted whole melons to gaseous ozone, hot water dipping and a combination of both treatments. Immersion in water at 75 °C for 1 min followed by treatment with 10 000 ppm gaseous ozone for 30 min resulted in 3.8, 5.1, 2.2 and 2.3 log reductions of mesophilic bacteria, psychrotrophic bacteria, yeasts/moulds and coliforms, respectively. When pre-cut melon cubes were used, Selma *et al.* (2008b) did not observe adverse effects on sensorial characteristics after treatment with gaseous ozone (at 20 000 ppm for 30 min) and during storage. Vurma *et al.* (2009) also tested combined treatments for *E. coli* inactivation in spinach. The combination of vacuum cooling (4 °C) with high gaseous ozone levels (935 ppm) for 30 min decreased the bacterium load by 1.8 log CFU/g, with no apparent damage of vegetable quality. However, when this combined treatment was sequentially followed by a treatment with low ozone levels (5–10 ppm) during 3 days, *E. coli* was undetectable (i.e. > 5 log reduction).

All data reported here showed that ozone treatments can be considered a suitable choice for low contaminated fruits and vegetables. However, optimization of ozone processing conditions must be assessed for a particular fruit and/or vegetable. The quality attribute mostly affected by ozone treatment is aroma, since several researchers observed reductions in aroma volatiles (Forney *et al.*, 2003, 2007; Perez *et al.*, 1999).

Residual pesticides are commonly found in fruits and vegetables, since significant quantities of these substances are often used for crop protection. Some pesticides, such as DDT (dichlorodiphenyltrichloroethane), diazinon and parathion generate concern due to their potential long-term adverse effects as carcinogenic substances (Bolognesi and Morasso, 2000). Several researchers determined the effectiveness of aqueous ozone in the degradation of several pesticides in fruits (Ong *et al.*, 1996) and vegetables (Wu *et al.*, 2007a, 2007b). However, care must be taken because ozonation of the pesticides may produce by-products potentially more toxic than the pesticides themselves (Ikehata and El-Din, 2005).

Several fruit juices have also been treated with ozone by bubbling the gas into the juices. In general, a desired 5 log microbial reduction can be achieved with ozone treatment. However, significant and undesirable changes in colour, anthocyanins and ascorbic acid contents are frequently observed.

Red meat, pork and poultry Undercooked meat is a common cause of foodborne illness due to its possible contamination with *E. coli* and *Salmonella* in beef and poultry, respectively. Therefore, several studies have been conducted to explore the efficacy of ozone treatment on red meat and poultry.

Some researchers applied aqueous ozone to beef carcasses, ground beef and beef surfaces as a method of improving the microbiological quality of the meat (Novak and Yuan, 2004a; Reagan *et al.*, 1996). Nevertheless, in the majority of the studies, it was concluded that reductions of target microorganisms are usually low because ozone is consumed by reacting with organic compounds present on the meat surface. Castillo *et al.* (2003) applied aqueous ozone (95 mg/L) to reduce the loads of *E. coli* and *S. typhimurium* on beef carcasses and verified that ozone treatment was equivalent to a water wash. Likewise, Novak and Yuan (2004b) concluded that aqueous ozone (5 ppm) had limited lethality against *Clostridium perfringens* vegetative cells and spores on beef surfaces. However, those authors enhanced microbial inactivation of ozone-treated meat by coupling with mild heat treatments.

Due to the limited efficacy of aqueous ozone treatments on meat products, several researchers studied the effect of ozonated water in combination with other antimicrobial methods, such as heat, NaCl or chlorine dioxide solutions (Novak and Yuan 2003). Although treatments using 1% aqueous ozone for 15 min combined with either cetylpyridinium chloride or acetic acid resulted in a slightly higher inactivation, these treatments affected the red colour of ground beef and the treatment using acetic acid produced off-odours (Pohlman *et al.*, 2002). The work of Kim and Shin (2011), however, suggested that ozonated water treatment (0.2 ppm for up to 60 min) effectively improved the chemical properties and safety of refrigerated meat.

Other investigators selected gaseous ozone for microbial control on meat. Cardenas *et al.* (2011) observed that the highest microbial inhibition was observed at 0 °C and after 24 h of exposure, producing a decrease of 0.7

and 2.0 log cycles in *E. coli* and total aerobic mesophilic microorganisms, respectively. However, the surface colour and lipid oxidation of the beef samples were unacceptable. Shorter exposure times (3 h) reduced 0.5 and 1.0 log cycles in the counts of total aerobic mesophilic microorganisms and *E. coli*, respectively, without changing the colour or producing rancidity in the beef.

Studies using gaseous ozone on pork meat with posterior vacuum packaging storage for up to 20 days at 4 °C established that 5–10 min of ozone exposure before packing may be a reasonable method regarding the effect of ozone level on meat oxidation, colour change and microbial reduction (Jeong *et al.*, 2007). Piachin and Trachoo (2011) also used pork meat to evaluate the effect of ozone gas and potassium lactate on lipid oxidation and *S. typhimurium* survival. Combinations of ozone and potassium lactate decreased only 1.5 log CFU/g of the target bacterium.

Chicken breasts were inoculated with *Salmonella infantis* or *Pseudomonas aeruginosa* and exposed to gaseous ozone (>2000 ppm for up to 30 min). Although no sensory deterioration was observed, microbial reductions were less than 2 log cycles (Al-Haddad, Al-Qassemi and Robinson, 2005). Trindade *et al.* (2012) compared ozone and chlorine solutions as sanitizing agents of chicken carcasses. They concluded that, in general, ozone was as effective as chlorine in the disinfection of chicken carcasses and can be a potential substitute of chlorine in poultry slaughter houses.

Ozone can also be used in the form of ozonated dry ice, applied for low-temperature food preservation during transport (Fratamico *et al.*, 2012). It was concluded that ozonated dry ice is a better alternative, especially for meat and poultry processors, when compared to dry ice without ozone content.

Fish The major pathogens associated with fish contamination are *Vibrio* spp. and *Aeromonas* spp., but concerning spoiling microflora, the most important microorganisms are *Pseudomonas* spp., *Shewanella* spp. and *Photobacterium* spp., which are distributed on skin, gills or in the gut and are able to grow during refrigerated storage (Cortesi *et al.*, 2009). Ozone has been studied at various stages of seafood processing and substantial data have been collected for shrimp, mussels and several varieties of fish. Meunpol, Lopinyosiri and Menasveta (2003) worked with black tiger shrimp and established that residual ozone concentrations of 0.35 mg/L in water for 30 min reduced *Vibrio harveyi* counts by approximately 3 log cycles. Chawla, Bell and Janes (2007) observed that soaking peeled shrimp meat in ozonated water was more effective than spraying shrimp with ozonated water and the higher ozone concentrations and longer treatment times studied were more effective for reducing levels of spoilage bacteria on the shrimp. They also verified that the application of aqueous ozone did not increase lipid oxidation in the shrimp immediately after treatment. Laboratory and pilot experiments were also

conducted using gaseous ozone to reduce *Vibrio* spp. bacteria in shrimp and results proved the treatment efficacy on microbial elimination (Blogoslawski and Stewart, 2011).

Ozone has also been applied as a sanitizer agent to extend the shelf-life of whole or filleted fishes. The combination of ozone with refrigerated storage prolongs the shelf-life of different fish species. Results of sensory analysis obtained by Gelman *et al.* (2005) showed that ozone treatment applied previously to storage at 0 °C of live tilapia prolonged the shelf-life by 12 days and improved quality characteristics. A new refrigeration system for fish storage was developed by combining an ozone generator with a slurry ice system and was applied to sardine and farmed turbot (Campos *et al.*, 2006, 2005). Results demonstrated that this system allowed a better maintenance of sensory and microbiological quality, which implied a shelf-life extension of 4 and 7 days for sardine and farmed turbot, respectively.

The combined effect of ozonated water and chitosan on the shelf-life extension of pacific oysters was studied by Rong *et al.* (2010). Based on microbiological analysis, biochemical indices and sensory evaluation, the researchers concluded that the combined treatment increased the shelf-life of the product from 8 to 20 days, indicating that ozonated water and chitosan have a great potential for oyster preservation.

Eggs Shell egg and egg-based products are one of the prime sources of salmonellosis (Mukhopadhyay and Ramaswamy, 2012). Several studies were conducted to evaluate ozone efficacy in decontamination of these products. In general, pressurized gaseous ozone has more success in microbial inactivation on shell eggs than aqueous ozone. Inoculated shell eggs dipped into ozonated water decreased the *Salmonella enteritidis* population by 2.5 log per egg (Koidis, Bori and Vareltzis, 2000).

With the objective of finding a time-saving, economical and effective egg sanitization treatment, Rodriguez-Romo and Yousef (2005) treated shell eggs externally contaminated with *S. enteritidis* with gaseous ozone, UV radiation and a combination of the two methods. Although the authors achieved 5.9 log reductions of *Salmonella* by applying ozone for 10 min, this treatment time is long for industrial application. The application of UV radiation followed by ozone provided a reduction of 4.6 log in only 2 min. Perry, Rodriguez-Romo and Yousef (2008) applied sequentially heat and ozone treatments to inactivate *S. enteritidis* on whole shell eggs and concluded that the combination was more effective than the processes applied individually.

Ozone may, however, negatively affect the egg components. This was the subject of the work of Fuhrmann *et al.* (2010), who applied gaseous ozone treatments to intact eggs and in separated egg components (egg white and egg yolk). Although significant alterations were observed after treatment with ozone (50 mL/L), at lower ozone doses the oxidative processes occurred

mainly at the egg surface, suggesting that ozone does not penetrate into the egg.

Due to the promising application of ozone treatments in eggs, there was a need to determine consumer acceptability of ozone-treated eggs. This was achieved by Kamotani *et al.* (2010), who reported that the overall visual and sensory acceptability of ozone-treated eggs were not significantly different from the ones used as control. Therefore, ozone-based processing can be a potential pasteurization technology for eggs, as it retains the quality characteristics important to consumers.

Dried foods Dried foods are highly susceptible to present fungal and bacterial spores due to their low water activity. As these spores are commonly resistant to antimicrobial treatments, ozone has been tested as a possible sanitizer of these products. Studies have been conducted in several dried foods, such as dried fruits and meat, cereal grains and spices.

The effect of ozone treatments on microbial decontamination of dried figs was widely investigated. When dried fruits were exposed to gaseous ozone, significant reductions of *E. coli*, *Bacillus cereus* and *B. cereus spores* were observed (Akbas and Ozdemir, 2008; Oztekin, Zorlugenc and Zorlugenc, 2006). Considerable changes in quality attributes were not detected. For peanut detoxification, it was concluded that ozone was effective in controlling aflatoxigenic fungi and also acted for the reduction of aflatoxin levels (de Alencar *et al.*, 2012). Ozone can also be used as a potential alternative to methyl bromide to treat dry-cured ham. It did not affect the overall quality and sensory characteristics when applied in the gaseous state at concentrations as high as 175 ppm during 48 h of exposure (Sekhon *et al.*, 2010).

The impact of ozone has also been evaluated for cereal grains, such as wheat, barley and rice. Allen, Wu and Doan (2003) verified that barley treated with gaseous ozone had a significant reduction of fungal spores and that ozone efficacy was enhanced by increasing the temperature and water activity. The same conclusions were achieved by Wu, Doan and Cuenca (2006) when gaseous ozone was used as a fungicide to preserve stored wheat. The effect of ozonation as a method to reduce *B. cereus* loads in processed rice was investigated by Shah *et al.* (2011). They observed reductions greater than 1.5 log cycles, revealing a significant impact on rice storage.

Dairy products Ozone has been applied to some dairy products, mainly milk and cheese. It was reported that ozone is effective in killing common dairy spoilage bacteria, such as *Pseudomonas fluorescens*, *P. fragi* and *P. putida*, after biofilm formation in milk (Dosti, Guzel-Seydim and Greene, 2005). Likewise, Greene, Few and Serafini (1993) concluded that ozonated water inactivated milk spoilage bacteria adhered in milk films on stainless steel. *L. monocytogenes* was also effectively inactivated by ozonation when inoculated in raw milk and in various branded milk samples (Sheela and

Muthukumar, 2011). However, the authors verified a negative effect of ozone on the nutritional content of samples, namely a decrease of 15% in protein and carbohydrate contents, and a reduction of around 10% in calcium content.

Ozone was also applied to enhance the ripening process of cheese and storage. Horváth, Bilitzky and Huttner (1985) reported an increase of 11 weeks in the storage period of cheese by the application of small ozone gas concentrations (0.02 mg/L) during the ripening period. The treatment was effective in destroying spores present on the cheese surface. Serra *et al.* (2003) used a cheese ripening room with a gaseous ozone atmosphere and evaluated the effectiveness of ozone on air quality and on the cheese surface. Results demonstrated that ozone was effective in reducing aerial fungal loads in the ripening room but not on the cheese surface.

Pastair, a Sweden-based dairy company, developed a cold pasteurization technique that uses ozone gas processors to produce safe cheese without damaging healthy components (ElAmin, 2007). The process consists of ozone injection into the product for a specific controlled time and then the product is heated to about 60 °C for a very limited time. This short time leads to a final product with minimal sensorial and nutritional changes.

25.5.2 Ozone as a disinfectant of surfaces and equipment

In any food processing facility, much attention is provided for cleaning and sanitation with the main goal of preventing food contamination. Substantial research has been done to evaluate the ozone action against different microorganisms on surfaces. It has been reported that ozone significantly decreases surface flora by at least 2 log cycles when applied both as a gas and in ozonated water (Lagrange, Reiprich and Hoffman, 2004; Moore, Griffith and Peters, 2000).

Pascual, Llorca and Canut (2007) gathered information about ozone as a surface disinfectant. They concluded that moderate ozone doses, between 0.5 and 3.5 ppm, are sufficient to achieve significant microbial inactivation, both in gas and aqueous form. However, when ozone is applied as a gas, the necessary exposure times are considerably longer (1–4 h) than when ozonated water is applied (1–10 min).

Cleaning and disinfection operations require high amounts of water and energy, generating wastewaters that have a negative environmental impact. Ozone saves water, because it does not leave residues and consequently does not require additional water rinses. As ozone does not need to be applied at high temperatures, its use also provides energy savings. Additionally, wastewaters can potentially be reused in the initial cleaning stages, either directly or after re-ozonation to attain the required quality (Pascual, Llorca and Canut, 2007).

Although the investment costs for ozonation systems are usually high, the lower maintenance and operation costs make the payback guaranteed after a short time. Due to this fact, ozone-based disinfection has been adopted mainly by the dairy, wine and brewing industries.

25.6 Remarks on health and safety concerns

Due to its strong oxidizing power, ozone is toxic above certain levels of concentration and length of exposure. At low concentrations, ozone is a respiratory irritant that can cause headaches, coughing, dizziness and nausea (Perry and Yousef, 2011). Exposure for long periods of time or to higher ozone concentrations can cause severe detrimental health effects and can even be fatal. Therefore, systems for ozone detection and destruction are crucial for worker safety in food processing facilities.

Several federal agencies have established health standards or recommendations to limit human exposure to ozone. In the United States, the Occupational Safety and Health Administration (OSHA) regulates ozone levels in the food industry. According to OSHA, workers must not be exposed to ozone concentrations higher than 0.1 ppm for extended periods of time or 0.2 ppm for short-term exposure (Occupational Safety and Health Administration, 2012).

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References

Achen, M. and Yousef, A.E. (2001) Efficacy of ozone against *Escherichia coli* O157:H7 on apples. *Journal of Food Science*, **66**(9), 1380–1384.

Akbas, M.Y. and Olmez, H. (2007) Effectiveness of organic acid, ozonated water and chlorine dippings on microbial reduction and storage quality of fresh-cut iceberg lettuce. *Journal of the Science of Food and Agriculture*, **87**(14), 2609–2616.

Akbas, M.Y. and Ozdemir, M. (2008) Application of gaseous ozone to control populations of *Escherichia coli*, *Bacillus cereus* and *Bacillus cereus* spores in dried figs. *Food Microbiology*, **25**(2), 386–391.

Al-Haddad, K.S.H., Al-Qassemi, R.A.S. and Robinson, R.K. (2005) The use of gaseous ozone and gas packaging to control populations of Salmonella infantis and Pseudomonas aeruginosa on the skin of chicken portions. *Food Control*, **16**(5), 405–410.

Allen, B., Wu, J.N. and Doan, H. (2003) Inactivation of fungi associated with barley grain by gaseous ozone. *Journal of Environmental Science and Health Part B – Pesticides Food Contaminants and Agricultural Wastes*, **38**(5), 617–630.

Baur, S., Klaiber, R., Hammes, W.P. and Carle, R. (2004) Sensory and microbiological quality of shredded, packaged iceberg lettuce as affected by pre-washing procedures with chlorinated and ozonated water. *Innovative Food Science and Emerging Technologies*, **5**(1), 45–55.

Bialka, K.L. and Demirci, A. (2007) Efficacy of aqueous ozone for the decontamination of *Escherichia coli* O157:H7 and *Salmonella* on raspberries and strawberries. *Journal of Food Protection*, **70**(5), 1088–1092.

Blogoslawski, W.J. and Stewart, M.E. (2011) Some ozone applications in seafood. *Ozone-Science and Engineering*, **33**(5), 368–373.

Bolognesi, C. and Morasso, G. (2000) Genotoxicity of pesticides: potential risk for consumers. *Trends in Food Science and Technology*, **11**(4–5), 182–187.

Campos, C.A., Rodriguez, O., Losada, V., Aubourg, S.P. and Barros-Velazquez, J. (2005) Effects of storage in ozonised slurry ice on the sensory and microbial quality of sardine (*Sardina pilchardus*). *International Journal of Food Microbiology*, **103**(2), 121–130.

Campos, C.A., Losada, V., Rodriguez, O., Aubourg, S.P. and Barros-Velazquez, J. (2006) Evaluation of an ozone-slurry ice combined refrigeration system for the storage of farmed turbot (*Psetta maxima*). *Food Chemistry*, **97**(2), 223–230.

Cardenas, F.C., Andres, S., Giannuzzi, L. and Zaritzky, N. (2011) Antimicrobial action and effects on beef quality attributes of a gaseous ozone treatment at refrigeration temperatures. *Food Control*, **22**(8), 1442–1447.

Castillo, A., McKenzie, K.S., Lucia, L.M. and Acuff, G.R. (2003) Ozone treatment for reduction of *Escherichia coli* O157:H7 and *Salmonella* serotype *typhimurium* on beef carcass surfaces. *Journal of Food Protection*, **66**(5), 775–779.

Chawla, A., Bell, J.W. and Janes, M.E. (2007) Optimization of ozonated water treatment of wild-caught and mechanically peeled shrimp meat. *Journal of Aquatic Food Product Technology*, **16**(2), 41–56.

Chiattone, P.V., Torres, L.M. and Zambiazi, R.C. (2008) Aplicação do ozônio na indústria de alimentos. *Alimentos e Nutrição, Araraquara*, **19**(3), 341–349.

Cho, M., Chung, H. and Yoon, J. (2002) Effect of pH and importance of ozone initiated radical reactions in inactivating *Bacillus subtilis* spore. *Ozone-Science and Engineering*, **24**(2), 145–150.

Cortesi, M.L., Panebianco, A., Giuffrida, A. and Anastasio, A. (2009) Innovations in seafood preservation and storage. *Veterinary Research Communications*, **33**, S15–S23.

Cullen, P.J. and Tiwari, B.K. (2012) Applications of ozone in fruit processing. In: Rodrigues, S. and Fernandes, F.A.N. (eds), *Advances in Fruit Processing Technologies*. CRC Press, Boca Raton, Florida, USA, pp. 185–202.

Cullen, P.J., Valdramidis, V.P., Tiwari, B.K., Patil, S., Bourke, P. and O'Donnell, C.P. (2010) Ozone processing for food preservation: an overview on fruit juice treatments. *Ozone-Science and Engineering*, **32**(3), 166–179.

de Alencar, E.R., Faroni, L.R.D., Soares, N.D.F., da Silva, W.A. and Carvalho, M.C.D. (2012) Efficacy of ozone as a fungicidal and detoxifying agent of aflatoxins in peanuts. *Journal of the Science of Food and Agriculture*, **92**(4), 899–905.

Demirkol, O., Cagri-Mehmetoglu, A., Qiang, Z.M., Ercal, N. and Adams, C. (2008) Impact of food disinfection on beneficial biothiol contents in strawberry. *Journal of Agricultural and Food Chemistry*, **56**(21), 10414–10421.

Dosti, B., Guzel-Seydim, Z. and Greene, A.K. (2005) Effectiveness of ozone, heat and chlorine for destroying common food spoilage bacteria in synthetic media and biofilms. *International Journal of Dairy Technology*, **58**(1), 19–24.

ElAmin, A. (2007) Dairy company tests ozone pasteurisation technique [online]. Available at: http://www.foodproductiondaily.com/Processing/Dairy-company-tests-ozone-pasteurisation-technique.

FDA (2001) Secondary Direct Food Additives Permitted in Food for Human Consumption, *Rules and Regulations,* Federal Register, vol. 66, no. 123, Sec. 173.368 Ozone.

FDA (2004) Juice HACCP: Hazards and Controls Guidance. *Guidance for Industry*, 1st edn.

Forney, C.F., Song, J., Fan, L.H., Hildebrand, P.D. and Jordan, M.A. (2003) Ozone and 1-methylcyclopropene alter the postharvest quality of broccoli. *Journal of the American Society for Horticultural Science*, **128**(3), 403–408.

Forney, C.F., Song, J., Hildebrand, P.D., Fan, L. and McRae, K.B. (2007) Interactive effects of ozone and 1-methylcyclopropene on decay resistance and quality of stored carrots. *Postharvest Biology and Technology*, **45**(3), 341–348.

Fratamico, P.M., Juneja, V., Annous, B.A., Rasanayagam, V., Sundar, M., Braithwaite, D. and Fisher, S. (2012) Application of ozonated dry ice (ALIGAL™ blue ice) for packaging and transport in the food industry. *Journal of Food Science*, **77**(5), M285–M291.

Freitas-Silva, O. and Venancio, A. (2010) Ozone applications to prevent and degrade mycotoxins: a review. *Drug Metabolism Reviews*, **42**(4), 612–620.

Fuhrmann, H., Rupp, N., Buchner, A. and Braun, P. (2010) The effect of gaseous ozone treatment on egg components. *Journal of the Science of Food and Agriculture*, **90**(4), 593–598.

Garcia, A., Mount, J.R. and Davidson, P.M. (2003) Ozone and chlorine treatment of minimally processed lettuce. *Journal of Food Science*, **68**(9), 2747–2751.

Gelman, A., Sachs, O., Khanin, Y., Drabkin, V. and Glatman, L. (2005) Effect of ozone pretreatment on fish storage life at low temperatures. *Journal of Food Protection*, **68**(4), 778–784.

Goncalves, A.A. (2009) Ozone – an emerging technology for the seafood industry. *Brazilian Archives of Biology and Technology*, **52**(6), 1527–1539.

Greene, A.K., Few, B.K. and Serafini, J.C. (1993) A comparison of ozonation and chlorination for the disinfection of stainless-steel surfaces. *Journal of Dairy Science*, **76**(11), 3617–3620.

Guzel-Seydim, Z., Bever, P.I. and Greene, A.K. (2004) Efficacy of ozone to reduce bacterial populations in the presence of food components. *Food Microbiology*, **21**(4), 475–479.

Guzel-Seydim, Z.B., Greene, A.K. and Seydim, A.C. (2004) Use of ozone in the food industry. *Lebensmittel-Wissenschaft und-Technologie (Food Science and Technology)*, **37**(4), 453–460.

Hall, R.M. and Sobsey, M.D. (1993) Inactivation of hepatitis-A virus and MS2 by ozone and ozone–hydrogen peroxide in buffered water. *Water Science and Technology*, **27**(3–4), 371–378.

Heim, C. and Glas, K. (2011) Ozone I: characteristics/generation/possible applications. *Brewing Science*, **64**, 8–12.

Herbold, K., Flehmig, B. and Botzenhart, K. (1989) Comparison of ozone inactivation, in flowing water, of hepatitis-A virus, poliovirus-1, and indicator organisms. *Applied and Environmental Microbiology*, **55**(11), 2949–2953.

Horváth, M., Bilitzky, L. and Huttner, J. (1985) *Ozone*. Elsevier, New York, USA.

Hunt, N.K. and Marinas, B.J. (1997) Kinetics of *Escherichia coli* inactivation with ozone. *Water Research*, **31**(6), 1355–1362.

Ijabadeniyi, O.A., Minnaar, A. and Buys, E.M. (2011) Effect of attachment time followed by chlorine washing on the survival of inoculated *Listeria monocytogenes* on tomatoes and spinach. *Journal of Food Quality*, **34**(2), 133–141.

Ikehata, K. and El-Din, M. G. (2005) Aqueous pesticide degradation by ozonation and ozone-based advanced oxidation processes: a review (Part I). *Ozone-Science and Engineering*, **27**(2), 83–114.

Jeong, J.Y., Kim, C.R., Kim, K.H., Moon, S.J., Kook, K. and Kang, S.N. (2007) Microbial and physiochemical characteristics of pork loin cuts treated with ozone gas during storage. *Korean Journal for Food Science of Animal Resources*, **27**(1), 80–86.

Kamotani, S., Hooker, N., Smith, S. and Lee, K. (2010) Consumer acceptance of ozone-treated whole shell eggs. *Journal of Food Science*, **75**(2), S103–S107.

Karaca, H. (2010) Use of ozone in the citrus industry. *Ozone-Science and Engineering*, **32**(2), 122–129.

Karaca, H. and Velioglu, Y.S. (2007) Ozone applications in fruit and vegetable processing. *Food Reviews International*, **23**(1), 91–106.

Khadre, M.A., Yousef, A.E. and Kim, J.G. (2001) Microbiological aspects of ozone applications in food: a review. *Journal of Food Science*, **66**(9), 1242–1252.

Kim, J.G. (1998) Ozone as an antimicrobial agent in minimally processed foods. Unpublished PhD Thesis, Ohio State University, USA.

Kim, C.K., Gentile, D.M. and Sproul, O.J. (1980) Mechanism of ozone inactivation of bacteriophage-f2. *Applied and Environmental Microbiology*, **39**(1), 210–218.

Kim, M.J. and Shin, H.S. (2011) Effect of treatment with ozonated water on shelf life of refrigerated meat. *Korean Journal for Food Science of Animal Resources*, **31**(4), 617–623.

Kim, J.G., Yousef, A.E. and Chism, G.W. (1999) Use of ozone to inactivate microorganisms on lettuce. *Journal of Food Safety*, **19**(1), 17–34.

Kim, J.G., Yousef, A.E. and Dave, S. (1999) Application of ozone for enhancing the microbiological safety and quality of foods: a review. *Journal of Food Protection*, **62**(9), 1071–1087.

Klingman, M.H. and Christy, A.D. (2000) Development of a continuous system for sanitizing whole apples with aqueous ozone. *Transactions of the ASAE*, **43**(6), 1989–1996.

Koidis, P., Bori, M. and Vareltzis, K. (2000) Efficacy of ozone treatment to eliminate *Salmonella enteritidis* from eggshell surface. *Archiv Fur Lebensmittelhygiene*, **51**(1), 4–6.

Komanapalli, I.R. and Lau, B.H.S. (1996) Ozone-induced damage of *Escherichia coli* K-12. *Applied Microbiology and Biotechnology*, **46**(5–6), 610–614.

Kowalski, W.J., Bahnfleth, W.P. and Whittam, T.S. (1998) Bactericidal effects of high airborne ozone concentrations on *Escherichia coli* and *Staphylococcus aureus*. *Ozone-Science and Engineering*, **20**(3), 205–221.

Lagrange, F., Reiprich, W. and Hoffmann, M. (2004) CIP-cleaning and disinfection with ozoned water. *Fleischwirtschaft*, **84**(2), 112–114.

Langlais, B., Reckhow, D.A. and Brink, D.R. (1991) *Ozone in Water Treatment: Application and Engineering*, Lewis Publishers, Chelsea.

Long, W., Pao, S., Inserra, P., Westbrook, E. and Ahn, S. (2011) Efficacy of ozone produce washers in reducing natural and artificially inoculated microorganisms on roma tomatoes and green onions. *Journal of Food Safety*, **31**(2), 268–275.

Mahapatra, A.K., Muthukumarappan, K. and Julson, J.L. (2005) Applications of ozone, bacteriocins and irradiation in food processing: a review. *Critical Reviews in Food Science and Nutrition*, **45**(6), 447–461.

Mahfoudh, A., Moisan, M., Seguin, J., Barbeau, J., Kabouzi, Y. and Keroack, D. (2010) Inactivation of vegetative and sporulated bacteria by dry gaseous ozone. *Ozone-Science and Engineering*, **32**(3), 180–198.

Meunpol, O., Lopinyosiri, K. and Menasveta, P. (2003) The effects of ozone and probiotics on the survival of black tiger shrimp (*Penaeus monodon*). *Aquaculture*, **220**(1–4), 437–448.

Miller, F.A., Silva, C.L.M. and Brandão, T.R.S. (2013) A review on ozone-based treatments for fruit and vegetables preservation. *Food Engineering Reviews*, **5**(1), 1–30.

Minas, I.S., Tanou, G., Belghazi, M., Job, D., Manganaris, G.A., Molassiotis, A. and Vasilakakis, M. (2012) Physiological and proteomic approaches to address the active role of ozone in kiwifruit post-harvest ripening. *Journal of Experimental Botany*, **63**(7), 2449–2464.

Moore, G., Griffith, C. and Peters, A. (2000) Bactericidal properties of ozone and its potential application as a terminal disinfectant. *Journal of Food Protection*, **63**(8), 1100–1106.

Mukhopadhyay, S. and Ramaswamy, R. (2012) Application of emerging technologies to control *Salmonella* in foods: a review. *Food Research International*, **45**(2), 666–677.

Muthukumarappan, K. (2012) Ozone processing. In: Sun, D.-W. (ed.), *Handbook of Food Safety Engineering*. Wiley-Blackwell, Oxford, pp. 681–692.

Novak, J.S. and Yuan, J.T.C. (2003) Viability of *Clostridium perfringens*, *Escherichia coli*, and *Listeria monocytogenes* surviving mild heat or aqueous ozone treatment on beef followed by heat, alkali, or salt stress. *Journal of Food Protection*, **66**(3), 382–389.

Novak, J.S. and Yuan, J.T.C. (2004a) The fate of *Clostridium perfringens* spores exposed to ozone and/or mild heat pretreatment on beef surfaces followed by modified atmosphere packaging. *Food Microbiology*, **21**(6), 667–673.

Novak, J.S. and Yuan, J.T.C. (2004b) Increased inactivation of ozone-treated *Clostridium perfringens* vegetative cells and spores on fabricated beef surfaces using mild heat. *Journal of Food Protection*, **67**(2), 342–346.

Occupational Safety and Health Administration (OHSA) (2012) Chemical sampling information – ozone, US Department of Labor [online]. Available at: http://www.osha.gov/dts/chemicalsampling/data/CH_259300.html.

O'Donnell, C., Tiwari, B.K., Cullen, P.J. and Rice, R.G. (2012) *Ozone in Food Processing*. Wiley-Blackwell, Oxford.

Olmez, H. and Akbas, M.Y. (2009) Optimization of ozone treatment of fresh-cut green leaf lettuce. *Journal of Food Engineering*, **90**(4), 487–494.

Ong, K.C., Cash, J.N., Zabik, M.J., Siddiq, M. and Jones, A.L. (1996) Chlorine and ozone washes for pesticide removal from apples and processed apple sauce. *Food Chemistry*, **55**(2), 153–160.

Oztekin, S., Zorlugenc, B. and Zorlugenc, F.K. (2006) Effects of ozone treatment on microflora of dried figs. *Journal of Food Engineering*, **75**(3), 396–399.

Palou, L., Smilanick, J.L., Crisosto, C.H. and Mansour, M. (2001) Effect of gaseous ozone exposure on the development of green and blue molds on cold stored citrus fruit. *Plant Disease*, **85**(6), 632–638.

Parish, M.E., Beuchat, L.R., Suslow, T.V., Harris, L.J., Garrett, E.H., Farber, J.N. and Busta, F.F. (2003) Methods to reduce/eliminate pathogens from fresh and fresh-cut produce. *Comprehensive Reviews in Food Science and Food Safety*, **2**, 161–173.

Pascual, A., Llorca, I. and Canut, A. (2007) Use of ozone in food industries for reducing the environmental impact of cleaning and disinfection activities. *Trends in Food Science and Technology*, **18**, S29–S35.

Patil, S. and Bourke, P. (2012) Ozone processing of fluid foods. In: Cullen, P.J., Tiwari, B.K. and Valdramidis, V.P. (eds), *Novel Thermal and Non-Thermal Technologies for Fluid Foods*. Academic Press, London, England, pp. 225–262.

Patil, S., Bourke, P., Frias, J.M., Tiwari, B.K. and Cullen, P.J. (2009a) Inactivation of *Escherichia coli* in orange juice using ozone. *Innovative Food Science and Emerging Technologies*, **10**(4), 551–557.

Patil, S., Cullen, P.J., Kelly, B., Frias, J.M. and Bourke, P. (2009b) Extrinsic control parameters for ozone inactivation of *Escherichia coli* using a bubble column. *Journal of Applied Microbiology*, **107**(3), 830–837.

Perez, A.G., Sanz, C., Rios, J.J., Olias, R. and Olias, J.M. (1999) Effects of ozone treatment on postharvest strawberry quality. *Journal of Agricultural and Food Chemistry*, **47**(4), 1652–1656.

Perry, J.J., Rodriguez-Romo, L.A. and Yousef, A.E. (2008) Inactivation of *Salmonella enterica* serovar enteritidis in shell eggs by sequential application of heat and ozone. *Letters in Applied Microbiology*, **46**(6), 620–625.

Perry, J.J. and Yousef, A.E. (2011) Decontamination of raw foods using ozone-based sanitization techniques. In: Doyle, M.P. and Klaenhammer, T.R. (eds). *Annual Review of Food Science and Technology*, vol. 2. Annual Reviews, Palo Alto, California, USA, pp. 281–298.

Piachin, T. and Trachoo, N. (2011) Effect of ozone and potassium lactate on lipid oxidation and survival of *Salmonella typhimurium* on fresh pork. *Pakistan Journal of Biological Sciences: PJBS*, **14**(3), 236–240.

Pohlman, F.W., Stivarius, M.R., McElyea, K.S., Johnson, Z.B. and Johnson, M.G. (2002) The effects of ozone, chlorine dioxide, cetylpyridinium chloride and trisodium phosphate as multiple antimicrobial interventions on microbiological, instrumental color, and sensory color and odor characteristics of ground beef. *Meat Science*, **61**(3), 307–313.

Rakness, K.L. (2005) *Ozone in Drinking Water Treatment: Process Design, Operation, and Optimization*. American Water Works Association, Denver, Colorado, USA.

Reagan, J.O., Acuff, G.R., Buege, D.R., Buyck, M.J., Dickson, J.S., Kastner, C.L., Marsden, J.L., Morgan, J.B., Nickelson, R., Smith, G.C. and Sofos, J.N. (1996) Trimming and washing of beef carcasses as a method of improving the microbiological quality of meat. *Journal of Food Protection*, **59**(7), 751–756.

Restaino, L., Frampton, E.W., Hemphill, J.B. and Palnikar, P. (1995) Efficacy of ozonated water against food-related microorganisms. *Applied and Environmental Microbiology*, **61**(9), 3471–3475.

Rey, R.P., Selles, A.N., Baluja, C. and Otero, M.L. (1995) Ozonation kinetics of glucosamine and *N*-acetyl glucosamine in aqueous-medium. *Ozone-Science and Engineering*, **17**(4), 463–467.

Rice, R.G., Farquhar, J.W. and Bollyky, L.J. (1982) Review of the applications of ozone for increasing storage times of perishable foods. *Ozone-Science and Engineering*, **4**(3), 147–163.

Rodgers, S.L., Cash, J.N., Siddiq, M. and Ryser, E.T. (2004) A comparison of different chemical sanitizers for inactivating *Escherichia coli* O157:H7 and *Listeria monocytogenes* in solution and on apples, lettuce, strawberries, and cantaloupe. *Journal of Food Protection*, **67**(4), 721–731.

Rodriguez-Romo, L.A. and Yousef, A.E. (2005) Inactivation of *Salmonella enterica* serovar enteritidis on shell eggs by ozone and UV radiation. *Journal of Food Protection*, **68**(4), 711–717.

Rong, C., Qi, L., Yin, B.Z. and Zhu, L.L. (2010) Combined effect of ozonated water and chitosan on the shelf-life of Pacific oyster (*Crassostrea gigas*). *Innovative Food Science and Emerging Technologies*, **11**(1), 108–112.

Sekhon, R.K., Schilling, M.W., Phillips, T.W., Aikins, R.M.J., Hasan, M.M., Nannapaneni, R. and Mikel, W.B. (2010) Effects of carbon dioxide and ozone treatments on the volatile composition and sensory quality of dry-cured ham. *Journal of Food Science*, **75**(5), C452–C458.

Selma, M.V., Ibanez, A.M., Allende, A., Cantwell, M. and Suslow, T. (2008a) Effect of gaseous ozone and hot water on microbial and sensory quality of cantaloupe and potential transference of *Escherichia coli* O157: H7 during cutting. *Food Microbiology*, **25**(1), 162–168.

Selma, M.V., Ibanez, A.M., Cantwell, M. and Suslow, T. (2008b) Reduction by gaseous ozone of *Salmonella* and microbial flora associated with fresh-cut cantaloupe. *Food Microbiology*, **25**(4), 558–565.

Serra, R., Abrunhosa, L., Kozakiewicz, Z., Venancio, A. and Lima, N. (2003) Use of ozone to reduce molds in a cheese ripening room. *Journal of Food Protection*, **66**(12), 2355–2358.

Shah, N., Rahman, R.A., Chuan, L.T. and Hashim, D.M. (2011) Application of gaseous ozone to inactivate *Bacillus cereus* in processed rice. *Journal of Food Process Engineering*, **34**(6), 2220–2232.

Sheela, M.M. and Muthukumar, M. (2011) Effectiveness of ozone in inactivating *Listeria monocytogenes* from milk samples. *World Journal of Life Sciences and Medical Research*, **1**(3), 40–44.

Shinriki, N., Ishizaki, K., Yoshizaki, T., Miura, K. and Ueda, T. (1988) Mechanism of inactivation of tobacco mosaic-virus with ozone. *Water Research*, **22**(7), 933–938.

Singh, N., Singh, R.K., Bhunia, A.K. and Stroshine, R.L. (2002) Efficacy of chlorine dioxide, ozone, and thyme essential oil or a sequential washing in killing *Escherichia coli* O157:H7 on lettuce and baby carrots. *Lebensmittel-Wissenschaft und-Technologie (Food Science and Technology)*, **35**(8), 720–729.

Thanomsub, B., Anupunpisit, V., Chanphetch, S., Watcharachaipong, T., Poonkhum, R. and Srisukonth, C. (2002) Effects of ozone treatment on cell growth and ultrastructural changes in bacteria. *Journal of General and Applied Microbiology*, **48**(4), 193–199.

Trindade, M.A., Kushida, M.M., Villanueva, N.D.M., Pereira, D.U.D. and de Oliveira, C.E.L. (2012) Comparison of ozone and chlorine in low concentrations as sanitizing agents of chicken carcasses in the water immersion chiller. *Journal of Food Protection*, **75**(6), 1139–1143.

Tzortzakis, N., Singleton, I. and Barnes, J. (2007) Deployment of low-level ozone-enrichment for the preservation of chilled fresh produce. *Postharvest Biology and Technology*, **43**(2), 261–270.

Vaduganathan, L., Poonamallie, B.A. and Nagalingam, M. (2012) Effects of temperature and flow rates of ozone generator on the DBD by varying various electrical parameters. *American Journal of Applied Sciences*, **9**(9), 1496–1502.

Venta, M.B., Broche, S.S.C., Torres, I.F., Perez, M.G., Lorenzo, E.V., Rodriguez, Y.R. and Cepero, S.M. (2010) Ozone application for postharvest disinfection of tomatoes. *Ozone-Science and Engineering*, **32**(5), 361–371.

Vurma, M., Pandit, R.B., Sastry, S.K. and Yousef, A.E. (2009) Inactivation of *Escherichia coli* O157:H7 and natural microbiota on spinach leaves using gaseous ozone during vacuum cooling and simulated transportation. *Journal of Food Protection*, **72**(7), 1538–1546.

Wei, K.J., Zhou, H.D., Zhou, T. and Gong, J.H. (2007) Comparison of aqueous ozone and chlorine as sanitizers in the food processing industry: impact on fresh agricultural produce quality. *Ozone-Science and Engineering*, **29**(2), 113–120.

Wu, J.N., Doan, H. and Cuenca, M.A. (2006) Investigation of gaseous ozone as an anti-fungal fumigant for stored wheat. *Journal of Chemical Technology and Biotechnology*, **81**(7), 1288–1293.

Wu, J.G., Luan, T.G., Lan, C.Y., Lo, T.W.H. and Chan, G.Y.S. (2007a) Removal of residual pesticides on vegetable using ozonated water. *Food Control*, **18**(5), 466–472.

Wu, J.G., Luan, T.G., Lan, C.Y., Lo, W.H. and Chan, G.Y.S. (2007b) Efficacy evaluation of low-concentration of ozonated water in removal of residual diazinon, parathion, methyl-parathion and cypermethrin on vegetable. *Journal of Food Engineering*, **79**(3), 803–809.

Young, S.B. and Setlow, P. (2004) Mechanisms of *Bacillus subtilis* spore resistance to and killing by aqueous ozone. *Journal of Applied Microbiology*, **96**(5), 1133–1142.

Yuk, H.G., Yoo, M.Y., Yoon, J.W., Moon, K.D., Marshall, D.L. and Oh, D.H. (2006) Effect of combined ozone and organic acid treatment for control of *Escherichia coli* O157:H7 and *Listeria monocytogenes* on lettuce. *Journal of Food Science*, **71**(3), M83–M87.

Zorlugenc, B., Zorlugenc, F.K., Oztekin, S. and Evliya, I.B. (2008) The influence of gaseous ozone and ozonated water on microbial flora and degradation of aflatoxin B(1) in dried figs. *Food and Chemical Toxicology*, **46**(12), 3593–3597.

26

Application of Pulsed Electric Fields in Food

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26.1 Introduction

Since the early 1960s and the pioneering steps made by German engineer Heinz Doevenspeck, pulsed electric fields (PEFs) appeared as a possibility for several different applications in food and bioprocessing. Nowadays, PEF is mostly being used in permeabilization of plant cells and for shelf life extension of heat sensitive liquid products. At present, more than 30 commercial units are operating worldwide. In 2006, the first commercial PEF application for juice preservation was installed in USA. Since then the interest for industrial scale processing equipment for liquid and solid products has increased. In 2009, the first industrial juice preservation line was installed in Europe, followed by the line for vegetable pre-treatment in 2010. The present chapter thus discusses the principle of PEF, commercial exploitation and equipment design.

As a part of the core business, it is necessary for food producers to optimize their processes throughout the production chain. Novel technologies allow the extension of the range of processes that are being used for separation, conversion, structuring or stabilizing food products. In the 1960s, PEF (sometimes also termed as electroporation or HELP – high intensity electric field pulses) application was proposed to induce cell disintegration in animal products to enhance mass transport and improve phase separation, as well as for microbial inactivation (Doevenspeck, 1960, 1961).

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First commercialization steps of the process were made in the 1980s, but at that time pulsed power switches had not shown sufficient performance and reliability (Sitzmann and Münch, 1988; Sitzmann, 2006). Nearly at the same time the impact of electroporation on plant, animal and microbial cells had been evaluated and numerous applications of electroporation in food and bioengineering had been investigated (Barbosa-Cánovas *et al.*, 1998; Toepfl, Heinz and Knorr, 2006). In the 1990s, in the USA as well as Europe, food processors, equipment manufacturers and universities formed consortia to develop PEF applications and equipment (Toepfl, Heinz and Knorr, 2006). In 1995, a continuous system was launched; in 2006, the first commercial installation for fruit juice preservation was implemented (Bushnell *et al.*, 1993), but was stopped in 2008 due to technical and commercial limitations. The first commercial application in Europe was realized in 2009, with the installation of a PEF unit with a capacity of 1.500 L/h for juice preservation. In 2010, the first industrial system for processing of vegetables with a maximum capacity of 50 t/h followed. Today, nearly 40 groups are working on PEF food processing and more than 500 research papers have been released related to the same subject. However, the aim of the process (level of cell disintegration or inactivation of certain microorganisms) as well as product parameters (e.g. pH, water activity, viscosity) contribute to the determination of the required processing conditions.

26.2 Principle of action

The application of an electric field to a cellular material induces permeabilization of the membrane. The membrane as a semi-permeable barrier for the intra- and extracellular transport of ions and macromolecules can be considered as a capacitor filled with dielectric material of low electrical conductance and a dielectric constant in the range of 2 (Castro, Barbosa-Cánovas and Swanson, 1993; Barbosa-Cánovas *et al.*, 1999a).

The exposure of the cell to an electrical field of a specific intensity causes a movement of free charges in and outside the cell along the electric field lines. This incidence results in the induction of additional potential. The transmembrane potential increases due to the electric field up to the value of 1 V (Zimmermann *et al.*, 1976; Weaver, 2000). The action of polarizing the cell is illustrated in Figure 26.1.

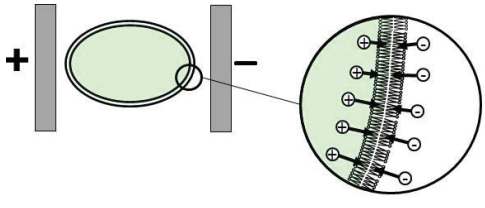

Figure 26.1 The action of cell polarization is illustrated in Saldaña (2011)

The polarization of the membrane is related to the increase of membrane conductivity in the range of more than 1 S/cm^2 (Dimitrov, 1984; Tsong, 1991; Wilhelm *et al.*, 1993). Hence, the resistance of the membrane decreases and results in a dramatic change in permeability for potassium and sodium ions (Pliquett *et al.*, 2007). The higher permeability leads to dielectric breakdown of the cell, which occurs at different electric potentials depending on the duration of the electrical pulse (Dimitrov, 1984).

Movement of free charges and the related polarization of the membrane result in the formation of electrocompressive forces, causing local dielectric rupture of the membrane and leading to the pore formation. New formed pores induce a drastic increase of permeability, termed as dielectric breakdown (Zimmermann *et al.*, 1976). If the applied electrical potential exceeds the critical potential, which depends on temperature, cell size and shape as well as medium and process conditions, the dielectric breakdown occurs (Dimitrov, 1984). The pore formation after PEF is illustrated in Figure 26.2 showing PEF treated yeast cells.

This electromechanical process is still the most accepted explanation for describing the effect of applying an external electric field on biological cells. Other theories describe the membrane as a viscoelastic model with fluctuating surfaces. The externally applied electric field induces reorientation and deterioration of the membrane molecules, resulting in pore formation and also the expansion of the pores and leading to mechanical breakdown of the membrane (Dimitrov, 1984).

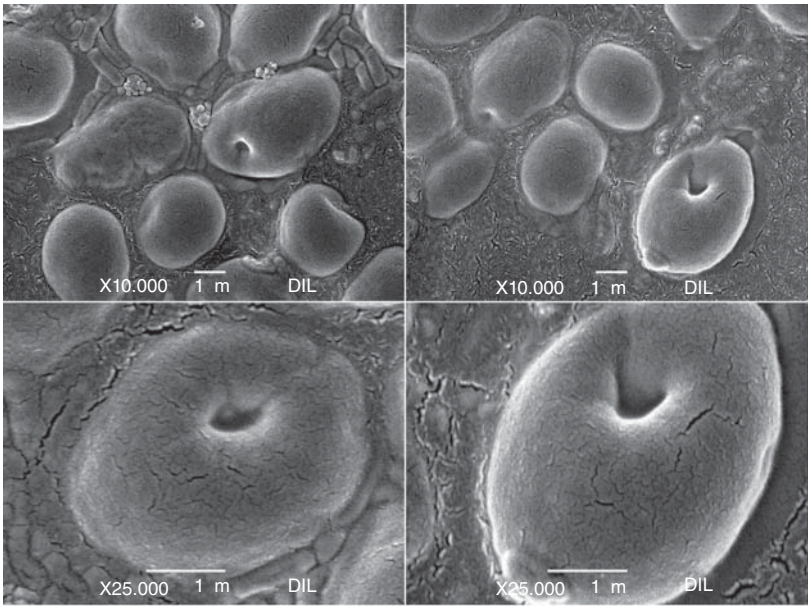

Figure 26.2 SEM exposure of yeast cells of *S. cerevisiae* (with different magnifications), treated by PEF (source: Aganovic *et al.*, 2012)

The created pores can vary in reversibility, depending on different intensities of the applied pulses. Exposing cellular material to pulses with less energy helps in the formation of reversible pores. Using a sufficient intensity of the electric pulses, stress reactions in plant cells can be induced and the production of secondary metabolites can be stimulated (Schilling *et al.*, 2007). Lethal cell damage is induced if the applied pulsed intensity exceeds the critical potential. This potential can be applied to inactivate pathogen and nonpathogen microorganisms or to facilitate the mass transfer, and also to improve the diffusion of intra- and extracellular liquids.

The application of PEF induces disintegration of plant/animal tissues (Heinz *et al.*, 2001; Toepfl, Heinz and Knorr, 2007) and microbial inactivation (Angersbach and Heinz, 1997; Angersbach, Heinz and Knorr, 2000). The differences between these two effects are due to applied intensity of the electric field and the specific energy. Those two parameters are considered as the main process parameters for PEF treatment. Electric field strength is defined as the electric potential difference for two given electrodes in the space divided by the distance between them, separated by a nonconductive material (Zhang, Barbosa-Cánovas and Swanson, 1995):

$$E = \frac{U}{d} \tag{26.1}$$

Here E is the electric field strength (kV/cm), U is the electric potential difference (V) and d is the distance between electrodes (cm).

The specific energy describes the intensity of the treatment with high intensity pulses and is expressed as energy in kJ per kg of the treated medium (kJ/kg). Because of the small size of the microorganisms and the composition of their membranes, a high energy input is required for a successful microbial inactivation (Glaser *et al.*, 1988), if compared to the energy level needed for cell disintegration. For microbial inactivation of different vegetative microorganisms, widely demonstrated by various researchers (Álvarez *et al.*, 2000; Toepfl, Heinz and Knorr, 2007), an electric field strength of around 15 to 20 kV/cm and a specific energy in the range of 40 to 1000 kJ/kg is required. In contrast, for the disintegration of plant cells, lower values for the electric field strength (0.7 to 3 kV/cm) and a specific energy (1 to 20 kJ/kg) are needed (Corrales *et al.*, 2008).

26.3 Application

Pulsed electric field technology induces a rupture of cell membranes and affects consequently all processes related to permeabilization of the cell. In the case of microbiological cells typical for food spoilage, the membrane is permeabilized, resulting in cell leakage. Thus, the spoilage of mainly beverages

can be avoided and shelf-life can be extended. The effect of PEF on plant material can also improve processes like extraction, distillation or drying.

26.3.1 Application in the juice industry

Apple juice The PEF treatment of apple juice, either fresh or made from concentrate, can extend shelf-life without affecting the quality of the juice. Treating the juice using PEF, different kind of microorganisms (e.g. *E. coli*, *S. cerevisiae*, *St. aureus* and *L. brevis*) can be inactivated (Akdemir Evrendilek *et al.*, 2000; García *et al.*, 2005a; Aguilar-Rosas *et al.*, 2007; Charles-Rodríguez *et al.*, 2007).

Garcia *et al.* (2005a) studied the effect of sublethal injury of *E. coli* after PEF treatment. For the treatment, square wave pulses with a pulse width of 2 μs and a frequency of 2 Hz were used. The results showed 1 log survival after the PEF treatment at 19 kV/cm and a treatment time of 400 μs. The use of selective agar medium showed 2 log sublethal injured *E. coli* cells after PEF treatment. Only 50% of the microbial cells were totally inactivated. During the storage at 4 °C, the sublethally injured cells were inactivated by the pH value of the medium. The same effect was detected by Garcia *et al.* (2005b). Applying 100 pulses with electric field strength of 30 kV/cm, survivor rate of 99% was detected. After 5 days a 5 log reduction of *E. coli* cells was obtained caused by the acid pH value.

Besides the microbial inactivation, the PEF impact on enzyme activity was investigated. Riener *et al.* (2008) studied the presence of polyphenoloxidase (PPO) and peroxidase (POD) after PEF treatment. Both enzymes catalyse oxidation reactions, leading to deterioration of colour, flavour and nutritional quality. Electric field strengths, applied in a batch chamber, were from 20 to 40 kV/cm, varying treatment times from 20 to 100 μs and inlet temperatures from 23 to 50 °C were investigated. The residual activity of the enzyme is mostly influenced by the inlet temperature prior to PEF. After the treatment (30 kV/cm, 100 μs treatment time), the relative activity of POD at 50 °C inlet temperature was 45%, compared to 63% at 30 °C. The same effect was detected for PPO (at 50 °C, the residual activity was 43% while at 23 °C, the residual activity was 59%).

Furthermore, the polyphenol content as a quality parameter responsible for colour and flavour development was analysed after PEF (bipolar pulses, 35 kV/cm) and thermal treatment (90 °C for 30 s). The studies showed a considerable loss of 14.5% polyphenols after PEF and 32.2% after thermal treatment (Aguilar-Rosas *et al.*, 2007).

Moreover, the dissipation of pesticides after PEF treatment was studied. Apple juice containing diazinon and dimethoate (the two common organophosphorus pesticides toxic to human health) was treated with exponential decay pulses with a pulse width of 10 μs. The intensity of the PEF

treatment varied by changing the electric field strength (8 to 20 kV/cm) and the treatment time (60 to 260 μs). The temperature did not exceed 23.5 °C. The results showed an increasing degradation of pesticides with increasing treatment time and electric field strength. After the treatment at 16 kV/cm and 190 μs, the residual concentration was 25.6%. The analysis of other organophosphorus pesticides, methamidophos and chlorpyrifos, studied by Chen *et al.* (2009), indicates the effect of PEF on their degradation. With increasing electric field strength and number of pulses, a higher reduction of pesticides was noticed. The persistent pesticide fractions of methamidophos and chlorpyrifos after PEF treatment with an electric field strength of 8 kV/cm for 26 pulses were 27.7 and 70.3%, respectively, which was lower than at 8 kV/cm for 6 pulses. A possible explanation for the degradation is based on the formation of hydrogen peroxide and hydroxyl radicals due to the electrode oxidation (Zhang *et al.*, 2012).

Volatile components responsible for the taste were examined by Aguilar-Rosas *et al.* (2007). The typical volatile compounds in apple juice were analysed after PEF treatment (35 kV/cm and 4 μs bipolar pulses) and compared to thermal treatment (90 °C for 30 s). The results of the analysis are shown in Table 26.1, which indicates a higher loss of volatile compounds after thermal treatment compared to PEF treatment. Nevertheless a loss of flavour was detected that occurred by PEF treatment.

The combination of PEF with other physical principles (high intensity light pulses (HILP) and UV light) enables the possibility for a gentle preservation of apple juice. Caminiti *et al.* (2011) studied the combined treatment of PEF and HILP at different intensities. For the HILP treatment, a xenon flash lamp was used with an energy level of 65.4 J/mL as a high treatment and 51.5 J/mL as a lower treatment. Energy input of 261.9 J/mL and field strength of 34 kV/cm were used for the high treatment and 130.5 J/mL and 24 kV/cm, respectively, for the lower PEF treatment. The highest inactivation of *E. coli* (>6.42 log reduction) was obtained using PEF treatment followed by HILP. After the treatments no changes in Brix, pH value or polyphenol content in

Table 26.1 Percentage loss of volatile compounds compared to an untreated sample after PEF and thermal treatment (source: Adapted from Aguilar-Rosas *et al.*, 2007. Reproduced with permission of Elsevier.)

	Loss after PEF (%)	Loss after thermal treatment (%)
Acetic acid	39.79 ± 20.84	100
Hexanal	7.04 ± 9.32	62.35 ± 5.35
Ethyl butyrate	60.19 ± 17.80	88.39 ± 12.46
Hexyl butyrate	8.41 ± 16.12	22.91 ± 21.99

the juice were observed. Moreover, no significant differences in colour, flavour or sweetness were also detected.

Besides HILP, PEF can be combined with UV light treatment. Noci *et al.* (2008) studied the effect of UV light at a wavelength of 254 nm for 30 min with 30 W intensity, combined with PEF treatment at an electric field strength of 40 kV/cm and 100 μs as the treatment time. Before the treatment, the juice was incubated for 48 h at 37 °C to increase the initial microbial count. The thermal treatment, characterized with a temperature at 94 or 72 °C for 26 s, caused a reduction of the total plate count of 6.7 and 6.0 log, respectively. The inactivation of UV treatment alone caused a reduction of 2.2 log cycles and PEF by 5.4 log cycles. A higher inactivation of 7.1 log was achieved combining PEF treatment and UV light. The quality of the juice was monitored by the pH values, °Brix, conductivity and polyphenol content, as well as enzyme activity (PPO, POD). The results showed a negligible effect of the combined treatment on the quality parameters. In the study conducted by Walkling-Ribeiro *et al.* (2008), the inactivation of *Staphylococcus aureus* was examined when combined treatment was applied. The UV light treatment caused an inactivation of 2.2 log cycles, whereas inactivation by PEF at 34 kV/cm for 63 μs treatment time was 5.8 log cycles. Combined treatments led to increased inactivation up to 8.3 log cycles. Increasing the inlet temperature and the electric field strength of the PEF treatment, a higher inactivation (9.3 log) could be achieved. However, pH, soluble solids and conductivity remained unaffected.

Orange juice Orange juice is a suitable product for the PEF treatment due to the presence of temperature-sensitive ingredients. The technology can offer an extended shelf-life while maintaining the fresh taste and quality of the juice. Min *et al.* (2003) studied the PEF treatment of orange juice on a commercial scale (500 L/h) to determine the shelf-life and juice quality. The total microbial count, and the yeast and mould counts were reduced to less than 10 CFU/mL after PEF treatment (electric field strength of 40 kV/cm and 2.6 ms treatment time). PEF treatment extended the shelf-life up to 196 days, compared to only 21 days of untreated fresh orange juice. McDonald *et al.* (2000) studied the inactivation of *E. coli*, *L. innocua*, *L. mesenteroides* and *S. cerevisiae* ascospores using PEF applying an electric field of 30 kV/cm and outlet temperature of 54 °C. This study showed 6 log reductions of *E. coli*, *L. innocua* and *L. mesenteroides*. The *S. cerevisiae* ascospores offered higher resistance to the PEF treatment. Selection of surrogate bacteria instead of *E. coli* O157:H7 and *Salmonella thyphimurium* were studied by Gurtler *et al.* (2010). Twenty-three strains were analysed in this study aiming on inactivation by PEF. The average inactivation of *E. coli* was 1.96 log (88.1% injury of survivors) using an electric field of 22 kV/cm, with an outlet temperature of 45 °C. An increase in the outlet temperature up to 55 °C caused an inactivation to 3.28 log, which represents 94.7% injury of the survivors. Analysing the *Salmonella* strains, higher resistance of the

virulence attenuated strains was detected. The study demonstrated different strain resistance to PEF treatment. Generally, the more intense treatments followed by a higher outlet temperature resulted in a higher inactivation rate of the analysed microorganisms.

Besides the energy input and other PEF parameters, the effectiveness of the PEF treatment is also influenced by microorganisms. The relation between the stage of inoculation and PEF treatment was studied by Molinari, Pilosof and Jagus (2004). Inactivation of *S. cerevisiae* using PEF in different growth stages was studied. Results showed a lower reduction in the logarithmic phase (3 log reductions), compared to the stationary phase (6 log reductions). The higher inactivation in the stationary phase is related to the inoculation level. Higher inoculation accelerates cell cluster formation, acting as one big cell, which in the end results in a higher inactivation using less energy.

PEF application enables an inactivation of spoilage microorganisms while maintaining the quality of the orange juice. The most important ingredient in orange juice is certainly L-ascorbic acid (vitamin C). After PEF treatment, a higher level of vitamin C concentration was detected compared to thermal treatment (Torregrosa *et al.*, 2006; Hodgins, Mittal and Griffiths, 2002). After PEF processing, the vitamin content was 55 mg/100 ml, which corresponds to the content of fresh orange juice. The content of the thermal treated juice was reduced to around 19% (Min *et al.*, 2003). During 196 days of juice storage, the vitamin C content of fresh juice, as well as PEF and thermal treated juices, decreased. However, for all analysed stages, the vitamin C content of fresh and PEF treated juice was higher than thermally processed juice (Min *et al.*, 2003). The bioavailability of vitamin C after PEF processing was studied by Sánchez-Moreno *et al.* (2005). A test panel drank 500 ml of the PEF treated juice two times a day and afterwards the plasma was analysed. The juice was processed in a continuous PEF system with a flow rate of 200 mL/min using an electric field strength of 35 kV/cm and treatment time of 750 μs. The results showed an increased ascorbic acid content in the plasma. Finally, the PEF processing retained the vitamin C bioavailability and antioxidant properties.

Other health-related significant components in orange juice are carotenoids. The study by Plaza *et al.* (2011) investigated the effect of PEF and high pressure (400 MPa, 1 min) and low-temperature pasteurization (70 °C for 30 s) on the carotenoid content of orange juice. The highest content was detectable after the high pressure treatment. No differences in carotenoid content after low pasteurization and PEF treatment were observed. No changes of pH value, Brix and colour were noticed after PEF treatment (Min *et al.*, 2003).

The antioxidant capacity of orange juice after PEF treatment was investigated and compared to thermal processing by Elez-Martínez and Martín-Belloso (2007). The effect of pulse polarity, shape and frequency was evaluated, as well as the electric field strength and treatment time. Higher retention of vitamin C was detected for bipolar pulses and lower electric field

strengths and frequencies as well as short pulse durations. The antioxidant capacity measured by the inhibition of 1,1 dipheny-2-picrylhydrazyl ($DPPH^{\cdot}$) radical indicated no differences between untreated or PEF processed juice.

The flavour of the juice after PEF treatment was examined by analysing the flavour generating compounds. The myrene volatile compound was retained (about 88%) after PEF treatment, compared to only 63% after thermal processing (Min *et al*., 2003). Other components, like ethylbutrate or decanal, indicated a retention of 97 and 100% after PEF treatment (30 kV/cm, 2 μs pulse duration, 480 μs treatment time) (Jia, Zhang and Min, 1999). In general, more flavour compounds were retained after PEF treatment compared to thermal treatment. The applied temperature for the thermal treatments in the studies of Min *et al.* (2003) and Jia, Zhang and Min (1999) was set to 90 °C.

The PEF treatment can be combined with other technologies to achieve a gentle preservation effect. Walkling-Ribeiro *et al.* (2009a) used PEF in combination with thermosonication. Before the PEF treatment (electric field strength of 40 kV/cm, 150 μs treatment time), the orange juice was sonicated at 55 °C for 10 min. No differences in colour, odour, acidity, sweetness, flavour and overall acceptability were detected. PEF not only can be used with other processes, but also it can be used in combination with antimicrobial substances. Hodgins, Mittal and Griffiths (2002) studied the presence of lysozyme and nisin on the microbial inactivation and the quality of orange juice. The results indicated synergistic effects between temperature, PEF and antimicrobial substances. Applying 20 pulses of 80 kV/cm, a 6 log inactivation can be achieved in the presence of 100 U Nisin/mL.

Smoothies and other juices Nowadays smoothies belong to a group of one of the most favourite beverages among consumers because of their health benefits. Most smoothies are squeezed and blended with fresh fruits, followed by direct packaging and going for retail in this form, having a shelf-life of only a few days. Using PEF technology, it is possible to extend the shelf-life, and preserve and maintain the fresh characteristics of smoothies, healthy ingredients and flavour compounds (Jin and Zhang, 1999; Cserhalmi *et al.*, 2006; Marsellés-Fontanet *et al.*, 2009; Walkling-Ribeiro *et al.*, 2009b).

Different types of smoothies were analysed, focusing on the effect of PEF on the nutritive ingredients. Morales-de la Peña *et al.* (2011) investigated the effect of PEF on smoothies made from orange, kiwi, pineapple, soymilk and sugar. PEF treatment applying the 35 kV/cm electric field strength and 1400 μs treatment time resulted in a 5.44 log reduction of *L. brevis* and 5.09 log of *L. innocua*. The shelf-life of the juice treated under the same conditions was 56 days, which is comparable to a thermally processed smoothie. No changes were observed on pH value and viscosity. The same effect was detected for vitamin C content; the samples were comparable to the untreated smoothie. Moreover, the PEF effect on fatty acids was analysed in the same study. Fatty acids such as palmitic, stearic, linoleic and oleic acids indicated no changes

after PEF and thermal processing. Other studies on grape juice indicated a marginal reduction of total fatty acids content of 51 mg/mL, compared to 54.1 mg/mL for untreated grape juice. Based on the disruption of the cells, some changes of amino acids were detected (Garde-Cerdán *et al.*, 2007).

A comparative study between high pressure (HP) and PEF processing, as well as with fresh (unheated) citrus juice (orange, grapefruit and tangerine juice) was done by Hartyáni *et al.* (2011) and Barba *et al.* (2012). No changes in soluble solid (°Brix), pH value, vitamin C content and conductivity were detected after PEF treatment using an electric field strength of 28 kV/cm for 100 μs treatment time, as well as after high pressure processing at 600 MPa for 10 min. For HP processed orange juice, colour change was detected. Moreover, the aroma and acid content of orange juice changed after HP treatment, whereas no effect of PEF on volatile flavour compounds of the juice was detected (Table 26.2). The analysis using electronic nose and tongue showed no differences between PEF or HP treatment and non-processed samples (Hartyáni *et al.*, 2011). The analysis of blueberry juice treated by HP and PEF resulted in a shelf-life of 56 days without quality loss (Aguiló-Aguayo *et al.*, 2009). A higher content of vitamin C was detected in HP treated juice (Barba *et al.*, 2012).

Besides the ingredients and the physical characteristics, enzymes play an important role in changing the structure, colour or physical properties of juices. In the study by Aguiló-Aguayo *et al.* (2008), the effect of PEF on lipoxygenase (LOX) and β-glucosidase (GLU) in strawberry juice has been reviewed. Process parameters like polarity, frequency and pulse width were studied, while an electric field strength of 35 kV/cm was held constant. Generally residual LOX activity, related to lipid oxidation, decreased with

Table 26.2 Organic acid content of different citrus juices after pulse electric field (PEF) and high pressure (HP) treatments compared to the control (source: Adapted from Hartyáni *et al.*, 2011. Reproduced with permission of Elsevier.)

Juice	Treatment	Malic acid (mg/L)	Citric acid (mg/L)	Ascorbic acid (mg/L)
Orange	Control	847.50 ± 70.06	5290.73 ± 207.48	511.59 ± 2.04
	PEF	826.24 ± 0.09	5222.57 ± 63.59	520.64 ± 12.93
	HP	755.77 ± 53.83	5207.17 ± 254.89	526.29 ± 17.64
Grapefruit	Control	537.42 ± 49.00	992.92 ± 80.86	421.18 ± 0.79
	PEF	494.71 ± 5.09	9833.22 ± 59.90	411.38 ± 6.96
	HP	452.75 ± 49.87	9751.39 ± 111.82	405.42 ± 5.56
Tangerine	Control	903.08 ± 112.20	341.41 ± 1.41	6318.03 ± 175.56
	PEF	944.02 ± 3.55	346.45 ± 1.15	6557.13 ± 7.03
	HP	1191.20 ± 105.92	386.49 ± 12.33	7596.88 ± 171.62

increasing frequency and pulse width. Enzyme GLU activity, responsible for colour and the key enzyme in flavour release, increased after PEF treatment (Aguiló-Aguayo *et al.*, 2008). The pectinmethylesterase (PME) and polygalacturonase (PG) were less affected by PEF treatment, compared to thermal treatment. The residual activity was about 22% (48% for thermal), resulting in the change of viscosity (Aguiló-Aguayo *et al.*, 2009). The same effect for the PME and PG was detected for smoothie types studied by Morales-de la Peña *et al.* (2011). Decreasing activity has been noticed with increasing treatment intensity.

The combination of PEF and antimicrobials seems to be a promising new approach to reduce the energy input required for a sufficient microbial inactivation. Mosqueda-Melgar, Raybaudi-Massilia and Martín-Belloso (2011) investigated the combined effect of PEF and antimicrobials like citric acid (CA) and cinnamon bark oil (CBO) on juices made from apple, pear, tomato, strawberry and orange, and compared the results with those obtained by thermal treatment. No changes of pH, conductivity and soluble solids could be detected (Table 26.3). Due to the combination of PEF with antimicrobials, the shelf-life was extended to 91 days. Although good results for microbiology and physical parameters were achieved, the sensory evaluation showed the lowest score in acceptability for the juice with added cinnamon bark oil (CBO). The citric acid (CA) also showed significant changes in terms of taste. Inoculation with *Salmonella enteriditis* and *E. coli* O157:H7 indicated additive and synergistic effects for most of the analysed juices, except in apple and pear juices. The antimicrobial citric acid showed a better inactivation

Table 26.3 Electrical conductivity, pH and soluble solids of pear and strawberry juices after PEF treatment in combination with antimicrobials compared to control and thermal treatment (source: Mosqueda-Melgar *et al.*, 2011. Reproduced with permission of Elsevier.)

Juice	Treatment	pH value	Conductivity (mS/cm)	Soluble solid content (%)
Pear	Control	4.85 ± 0.01	3.04 ± 0.03	15.4 ± 0.1
	PEF	4.87 ± 0.02	3.01 ± 0.04	15.3 ± 0.2
	PEF + CA	2.91 ± 0.01	3.15 ± 0.02	16.1 ± 0.2
	PEF + CBO	4.85 ± 0.01	$2,99 \pm 0.02$	15.4 ± 0.1
	Thermal	4.84 ± 0.03	$3,04 \pm 0.02$	15.6 ± 0.1
Strawberry	Control	3.26 ± 0.01	3.78 ± 0.08	7.0 ± 0.1
	PEF	3.27 ± 0.02	3.80 ± 0.06	7.0 ± 0.1
	PEF + CA	2.93 ± 0.02	3.92 ± 0.01	7.3 ± 0.1
	PEF + CBO	3.4 ± 0.01	3.81 ± 0.04	7.1 ± 0.1
	Thermal	3.25 ± 0.02	3.91 ± 0.02	7.4 ± 0.2

CA, citric acid; CBO, cinnamon bark oil.

effect than cinnamon bark oil (Mosqueda-Melgar, Raybaudi-Massilia and Martín-Belloso, 2008a, 2008b, 2011).

Besides citric acid and cinnamon bark oil, nisin can be also used as an antimicrobial agent. Nguyen and Mittal (2007) investigated the effect of nisin (100 U/mL) in combination with PEF in tomato juice. Synergistic effects were detected for the inactivation of the total plate count. Inactivation was increased up to 4.4 log for the combined treatment, compared to 0.85 log for the nisin application alone and 1.4 log just for the PEF treatment. In combination with both, a shelf-life of 28 days was achieved. A combination of different antimicrobials, for example nisin and lysozyme, has shown further improvement in inactivation in grape juice (Wu, Mittal and Griffiths, 2005).

Gurtler *et al.* (2011) studied the effect of PEF in combination with sodium benzoate (750 ppm) and potassium sorbate (350 ppm), comparing additionally the impact of citric acid. Different outlet temperatures after PEF treatment were selected to analyse the effect of temperature. *E. coli* O157:H7 and a nonpathogenic surrogate *E. coli* were chosen as the target microbes. The results (Table 26.4) indicate increased inactivation with increasing outlet temperature. A synergism between PEF and antimicrobials was not observed. Lowering the pH value by adding citric acid resulted in increased inactivation of both analysed *E. coli* strains.

Inactivation of spores by PEF So far the inactivation of vegetative microorganisms by PEF was commonly studied. Bacterial endospores showed higher resistance to temperature, presence of chemicals, extreme environmental conditions and also to PEF treatment (Setlow *et al.*, 2006; Yonemoto *et al.*, 1992).

Table 26.4 Inactivation of pathogen and nonpathogen *E. coli* strains after PEF treatment in combination with antimicrobials (source: Adapted from Gurtler *et al.*, 2011. Reproduced with permission of Elservier.)

		Inactivation (log N/N_0) at different outlet temperatures (°C)		
		45	50	55
E. coli 0157:H7	PEF	3.09	4.08	4.71
	PEF + antimicrobials	2.27	3.29	5.40
	PEF + antimicrobials + citric acid	2.60	4.32	6.95
Nonpathogen *E. coli*	PEF	2.80	3.12	3.79
	PEF + antimicrobials	2.75	3.52	5.11
	PEF + antimicrobials + citric acid	3.54	5.69	7.13

Thermal sterilization requires high temperatures of more than 100 °C and few minutes of holding time, depending on the targeted spore (Kessler, 1996). Using PEF in combination with thermal energy, bacterial endospores can be inactivated at a lower temperature compared to the thermal sterilization process (Siemer *et al.*, 2014).

Several studies indicated no inactivation of spores using temperatures lower than 60 °C and electric field strengths less than 30 kV/cm (Hamilton and Sale, 1967; Knorr *et al.*, 1994; Pagán *et al.*, 1998; Barbosa-Cánovas *et al.*, 1999b; Cserhalmi *et al.*, 2002; Shin *et al.*, 2008). Increasing the temperature and electric field strength, an inactivation of, for example, *B. subtilis* or *B. cereus* could be achieved (Marquez, Mittal and Griffiths, 1997; Siemer *et al.*, 2011; Bermúdez-Aguirre, Dunne and Barbosa-Cánovas, 2012). The study of Siemer *et al.* (2011) showed the inactivation of *B. subtilis* spores in Ringer solution with different conductivities and pH values by PEF in combination with thermal energy. Before the PEF treatment, Ringer's solution inoculated with spores was preheated up to 80 °C. As a result, complete inactivation of spores was detected in PEF treated samples. Higher medium conductivities require less energy to achieve the maximum inactivation. The pH value influences the inactivation of *B. subtilis* spores, enabling a lower energy input for the same level of inactivation at lower pH values.

Ohmic heating is another manner of thermal treatment, where the heat is generated by applying electrical current through food as a conductor. It shows better inactivation of spores compared to the traditional thermal sterilization process. Furthermore, the retention of quality parameters, such as ascorbic acid, was higher after ohmic heating (Cho, Yousef and Sastry, 1999; Uemura and Isobe, 2003). Synergistic effects of the temperature and electric field were noticed. The other combined treatments, like high pressure and PEF, resulted in higher inactivation compared to each treatment alone (Spilimbergo *et al.*, 2002).

26.3.2 Cell disintegration by PEF

Mass transfer processes are important for many industrial productions, like:

- Dehydration/freezing
- Extraction
- Distillation.

Drying process Removal of water from food can be described as a preservation method by lowering the water activity. Drying can also be used as a pre-treatment for improving the nutritional, organoleptic and functional values of food products (Torreggiani, 1993). The most important factor during the drying process is the force needed to remove water. This force can be described as a cause of mass transfer and is one of the major process steps

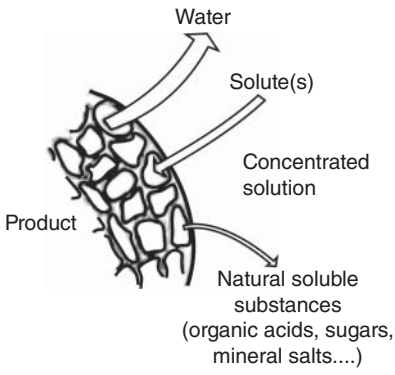

Figure 26.3 Mass transfer during the osmotic dehydration (OD) process (source: Adapted from Torreggiani, 1993. Reproduced with permission of Elsevier)

together with momentum and heat transfer during the drying process. In general, drying is based on the equilibrium status of the moisture content. The food loses or gains moisture over a period of time to attain a new equilibrium status (Sharma, Mulvaney and Rizvi, 2000).

Osmotic dehydration (OD) is a drying process possibly suitable for heat-sensitive products; it can be used as a pre-treatment to reduce the drying time (Ade-Omowaye *et al.*, 2001). The food is placed in an osmotic solution resulting in the formation of water and solubility gradient across the cell membrane. The membrane separates the cell content, mainly water and the osmotic solution. Due to the gradient, two fluxes are formed: water out of the cell and osmotic solution into the cell. This flux is a mass transfer process and is a function of the difference in chemical potentials. Figure 26.3 illustrates the flux of water and solute in and out of the cell (Torregiani, 1993; Sharma, Mulvaney and Rizvi, 2000).

The PEF usage as a pre-treatment affects the mass transfer and therefore the OD process at the same time. Facilitated mass transfer during OD using PEF was reported by Rastogi, Eshtiagi and Knorr (1999) and Ade-Omowaye *et al.* (2001). Selective permeabilization of the cell membrane allows easy and fast exit of the water in the osmotic solution, but no solid uptake (Taiwo *et al.*, 2003). Using PEF before the OD process, the drying time can be significantly reduced and a better structured food quality can be obtained.

The effect of PEF on the drying time within an OD process was studied by Ade-Omowaye *et al.* (2001, 2003). PEF treatment of red pepper indicated facilitated moisture removal and improved quality, especially in terms of colour. After 1.5 h of drying at 60 °C, the moisture content of the untreated red pepper was 2.06 kg/kg. Applying the PEF with an electric field strength of 1 and 2 kV/cm, the moisture content can be reduced to 1.45 and 1.09 kg/kg, respectively.

	Cutting force [N/cm^2]	
	Mark	Rind
Untreated	2.39 ± 0.62	2.07 ± 0.64
Thermal	1.06 ± 0.15	0.73 ± 0.02
PEF	0.47 ± 0.02	0.49 ± 0.04

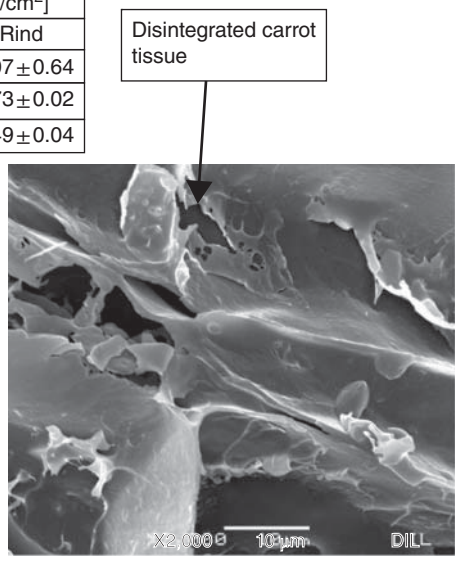

Figure 26.4 Cutting force and microscopic pictures from carrot tissue after PEF treatment compared to control and thermal treatment (80 °C for 20 min) (source: Adapted from Siemer *et al.*, 2012)

PEF can also be used for pre-treatment of carrots in order to reduce the OD time from 4 to 2 h by increasing the diffusion coefficient (Rastogi, Eshtiagi and Knorr, 1999). The PEF-induced enhanced mass transfer depends on the electric field strength and the applied number of pulses. The improved mass transfer is based on the permeabilization of cells induced by PEF and resulted in a softer tissue structure characterized by the lower cutting force (Figure 26.4) (Siemer *et al.*, 2012). Amami *et al.* (2007a, 2007b) studied PEF impact on sliced carrots. They reported that the electric field of 0.6 kV/cm and total energy input of 19 kJ/kg resulted in less water loss after the PEF process compared to the static OD process. Water loss after the static OD process was around 42% and water loss involving the PEF process was 38%. The same effect of the improved OD process was reported for apple tissue (Amami, Vorobiev and Kechaou, 2006; Chalermchat, Malangone and Dejmek, 2010) and for strawberries (Taiwo *et al.*, 2003).

Another potential application of PEF treatment was aimed to reduce the drying or freezing time of potatoes. Lebovka, Shynkaryk and Vorobiev (2007) studied freeze drying of PEF-treated potato slices and reported less structural damage. This appearance can be explained by ice formation during the freezing process. Facilitated water removal induces the ice formation and starts first at the outside the cell (Jalté *et al.*, 2009).

Not only drying process but also freezing process is recognized as a mass transport dependent process. Suitable products for this type of treatment are

fruits (strawberries and raspberries), vegetables (peas and green beans) as well as fish and meat products (Sharma, Mulvaney and Rizvi, 2000). The process is based on a decrease of temperature under the freezing point and combining the effect of low temperature with the conversion of water into ice (Delgado and Sun, 2001).

The time required to reach the freezing temperature is called the freezing time. This time can be predicted with mathematic modelling under the combination of heat and mass transfer. Therefore, the heat and mass transfer phenomena have to be considered. The application of PEF improves the mass transfer. The pre-treatment of potatoes (Jalté *et al.*, 2009) induces a higher freezing rate with a shorter freezing time. Due to PEF treatment, the cell membrane becomes porous, resulting in an enhanced diffusion controlled mass exchange of extracellular and intracellular water as well as a reduced freezing time. PEF thus acts as a pre-treatment for achieving a better quality of frozen food as smaller ice crystals are formed. The structure and form of the PEF pre-treated potato was better in comparison to the untreated samples.

Extraction Besides drying and freezing, PEF treatment influences the extraction process. In general, extraction is a process of separating a substance from a matrix. The aim of extraction process is the separation of desired extract located in the solid (mostly in the pore or cell structure of the solid) by using a solvent in which the extract is soluble. This technique has been used for a long time for the extraction of plant components, for example sucrose from sugar beet or tannin or colour extracts. The solid-liquid extraction is mostly used for (Bouzrara and Vorobiev, 2003):

- Extraction of fruit juices and vegetable oils
- Production of wine
- Dewatering of fibrous materials (sugar beet)
- Dehydration of organic wastes.

The principle of the extraction process is based on the mass transfer of the extract from the solid in solvent. The extract is mostly located in cells, which are surrounded by membranes. Consequently, the degree of damaged cellular material is related to the amount of released compounds. The effect of PEF can be defined as electroporation resulting in a disintegration of cellular material and an improved mass transfer. Regarding the extraction process, a facilitated extraction is possible using PEF as a pre-treatment, because the extract can easily diffuse into the solvent. Using PEF as a pre-treatment, the extraction time can be reduced and the yield of important quality ingredients can be increased.

The use of PEF for an improved extraction process can be applied in different fields. One example is the extraction of calcium from bones (Yin and He, 2008). There are different methods used for this extraction, like boiling

or microwave treatment, but they often affect the product negatively and the extracted concentration is low. Using PEF, the temperature can be reduced while the calcium concentration can be increased (Yin and He, 2008).

Another example for a solid-liquid extraction assisted by PEF treatment is the sugar beet. Traditionally, the cossettes are exposed to a temperature of 70 to 75 °C for 1 to 1.5 h to enhance the sucrose extraction. Applying 20 pulses with an electric field strength of 7 kV/cm, temperature can be reduced from 70 °C down to 40 °C in a 60 min extraction process, where the sucrose yield of 80% could be obtained. Using PEF, the energy consumption can be reduced to 50% (López *et al.*, 2009). Thermal treatment at 70 °C leads to sucrose concentration of 14% with a purity of more than 90%. The same values can be achieved by applying the PEF at a temperature of 30 °C (Loginova, Lebovka and Vorobiev, 2011). The combination of PEF and agitation can also lead to an increase in extraction yield. El-Belghiti and Vorobiev (2004) reported 30% extraction yield achieved by including agitation (100 rpm) after the PEF process (0.9 kV/cm, 250 pulses) within 120 min, compared to 500 min without agitation. The extraction yield can also be improved up to 20% by applying pressure of 30 MPa (Eshtiaghi and Knorr, 2002).

Lower extraction temperatures during the PEF process offers an increase in the extraction yield and better colourant retention at the same time. The major colourant in beet root is the temperature-sensitive betalains. Thermal treatment at 80 °C for 1 h induces complete degradation of the colourant. Using PEF at 30 °C with an electric field strength of 1.5 kV/cm for 20 pulses, the same extraction yield can be achieved as for thermal treatment at 80 °C for 40 min, though less colour degradation was observed (Fincan, DeVito and Dejmek, 2004; Loginova, Lebovka and Vorobiev, 2011).

Increased juice yield is of great importance from the economic aspect. Most commonly employed mechanical, enzymatic or temperature-based methods can be used for cell rupture for a facilitated extraction. However, they often lead to a degradation of important juice components as well as higher energy consumption (Bazhal and Vorobiev, 2000; Bazhal, Lebovka and Vorobiev, 2001; Toepfl, 2006; Toepfl and Heinz, 2011). PEF treatment with an electric field strength of 1 and 5 kV/cm and energy input of 1 to 30 kJ/kg increased the yield in the range of 1.7 to 7.7% in comparison to an enzymatic treatment (4.2%) (Schilling *et al.*, 2007). Moreover, the pomace can be used after the treatment for pectin extraction.

Grimi *et al.* (2011) reported a higher polyphenol content after PEF pre-treatment compared to untreated apple juice. Similar results of increased polyphenol content after PEF pre-treatment have been observed in wine production. The polyphenols responsible for the colour and the flavour of wine are located in the grape skin. By rupturing the skin, a facilitated polyphenol extraction can be achieved (Corrales *et al.*, 2008; Puértolas *et al.*, 2009, 2010). PEF-induced cell disintegration using an electric field strength of 3 kV/cm and 10 kJ/kg energy resulted in an increase in the phenolic compounds as

well as a higher antioxidant content (7841 μmolTE/g DM in comparison to 187 μmolTE/g DM thermal treatment at 70 °C for 1 h) (Corrales *et al.*, 2008).

Moreover, oil extraction can be improved using PEF (Guderjan *et al.*, 2005; Guderjan, 2006; Guderjan, Elez-Martinez and Knorr, 2007). The treatment of maize at 20 °C with an electric field strength of 0.6 kV/cm and specific energy of 0.62 kJ/kg led to an increase of oil yield up to 43.7% compared to 23.2% after thermal treatment. The same effect could be observed for PEF treatment of olives. Treating olives with 100 pulses of an intensity of 1.3 kV/cm can increase the yield up to 7.4% (Guderjan *et al.*, 2005).

Distillation Another process limited by mass transfer is the distillation process. In this process, a high temperature is applied to the product for a defined time in order to separate two components with different boiling points. Due to the high temperature, the cellular tissue is denatured and a facilitated extraction of the product can be achieved. At the same time, a higher heat load degrades the heat-sensitive ingredients, causing a quality loss of the product. The membrane of the cells represents a semi-permeable barrier that is impermeable for the desired extract due to its size. If the cells are ruptured, the extract could easily diffuse along the concentration gradient. Another possibility to rupture the cell tissue without any heat load, and consequently without any heat-related quality loss, is the application of PEF. In addition, the extraction yield could also be increased.

PEF treatment of roses is possible to gain a higher yield of rose oil while reducing the distillation time (Dobreva *et al.*, 2010). After a distillation time of 1 h, the same extraction yield was observed for PEF pre-treated roses as for the control, including 2.5 h of distillation time. Using a PEF treatment of roses with an electric field strength of 4 kV/cm and a specific energy of 10 kJ/kg, the yield can be increased up to 35% compared to the traditional thermal process. By reducing the extraction time, the energy consumption can be lowered and higher productivity of the process can be achieved.

26.4 Equipment design

Since the first promising trials on PEF, the equipment design and related scalability have been brought into focus in the industry. Basically a PEF system is built up of a pulsed power supply and a treatment chamber (Gaudreau *et al.*, 2004; Loeffler, 2006; Gusbeth *et al.*, 2009). Up to the 1990s, mostly spark gaps and vacuum tubes had been applied for power switching, showing poor reliability and a short lifetime (Sitzmann, 2006). The significant progress in the high-power semiconductor industry was achieved due to the high demand in medical, defence and energy-producing industries. However, the use of high-performance thyristors or transistors, which are required for a PEF system in the food industry, is still challenging. The environmental and operating

conditions may cause high maintenance efforts and risk of failure (Toepfl, Heinz and Knorr, 2006). The use of custom-designed, single-pulsed power switches may result in equipment downtime and idle times until replacement. Recent developments therefore have focused on modular designs to allow the use of standard components and to reduce maintenance (Toepfl, 2011). The design of PEF systems focuses on hygienic design and also an easy implementation in existing HACCP concepts. Furthermore, the safety and operation of the equipments are of importance, as often untrained operators are employed at the shop-floor level.

Different setups of pulse transformer or semiconductor-based Marx generators have been developed for switch devices in PEF equipments (Kuthi *et al.*, 2003; Kern, 2005; Toepfl, 2011). In the case of the pulse transformer, capacitors are charged by the power supply and the pulsed current rate is controlled by transistors. By using different arrangements, the intermediate circuit voltage can be changed in order to change the polarity. Switching the transistor to the 'on' position, the voltage is transformed to a desired value. Within this system, only a limited amount of capacity and related peak power can be achieved. The equipment developed by German Institute of Food Technologies (DIL e.V.) consisting of a semiconductor-based Marx generator can operate at a higher peak power and bigger capacities can be achieved. This principle is based on an installed stack of capacitors, which are charged in parallel. The discharge of the capacitors is controlled by the transistors and happens in series. Due to this setup a voltage multiplication can be achieved. Furthermore, in the case of a breakdown of a stack, the unit is still operating. Figure 26.5 shows the two realized units.

Figure 26.5 PEF units developed by DIL and the related maximum capacities; left side: pulse transformer setup; right side: semiconductor-based Marx generator equipment (source: DIL e.V., Quakenbrueck, Germany). *See plate section for colour version*

Though from an application point of view the average power level defines the maximum treatment capacity, in most cases the peak power has been shown to be the major challenge and cost driver. The typical average power of today's PEF units is in the range of 30 to 400 kW (Schultheiss *et al.*, 2004; Loeffler, 2006; Ravishankar, Zhang and Kempkes, 2008). The peak power can range up to several hundred megawatts, in particular when using treatment chambers with a large cross-section for solid products.

The PEF units developed by DIL can be used for microbial inactivation as well as cell disintegration. For a fluid pumpable product's pipe systems, varying in size according to the required capacity and product characteristics are available. For the treatment of solid materials, such as potatoes or carrots, a belt PEF system can be used. Different capacities can be obtained by varying the size of the belt.

26.5 Outlook

Novel technologies have been developed to improve food processing with regards to energy and time saving as well as quality improvement. Pulsed electric field (PEF) technologies as a novel technology offer the possibility of producing food possessing a high-quality level.

By applying PEF, the membrane of microbial cells as well as cellular tissue is permeabilized. In the case of permeabilization of microbial cells, a positive effect on the inactivation and shelf-life could be detected. Studies analysing the shelf-life of PEF-treated juices compared to untreated juices showed an extended shelf-life. Besides the microbial safety, the quality parameters, for example vitamin C content, Brix or colour, were analysed before and after PEF treatment, as well as compared to thermal treatment. In most of the cases, the PEF-treated samples were reported as the products with superior quality attributes compared to their thermal counterparts. Certain studies analysed the effect of PEF combined with antimicrobial substances or nonthermal processes. Further studies are required aiming at scalability and basic studies are needed to understand the combined effect. In addition, investigations are to be focused related to the process of sterilization by PEF. In the past, studies showed no effect of PEF on bacterial endospores. Within the development of new equipment designs and also the combination with thermal energy, the inactivation of spores can be achieved. At present only a few studies are available indicating a reduction of spore count of different species in various products. More research is required in the field of the principle of inactivation of spores by PEF as well as basic studies aiming at studying the influence of process and product parameters.

An improved mass transfer, necessary for the extraction or drying processes, can be achieved by PEF treatment of cellular tissues. In several studies, the PEF treatment of various products, for example potatoes, have

shown the influence of process parameters. These studies show an improved production process with regard to conservation of energy and time, resulting in a more efficient and economic production. Due to the development of PEF equipments on a high capacity as well as connection to a belt system, PEF treatment is aiming at cell disintegration, which is now the focus of different manufacturers. In summary, the application of PEF improves food production in order to enable the production of high-quality food in an efficient manner.

References

Ade-Omowaye, B.I.O., Angersbach, A., Taiwo, K.A. and Knorr, D. (2001) Use of pulsed electric field pre-treatment to improve dehydration characteristics of plant based foods. *Trends in Food Science and Technology*, **12**, 285–295.

Ade-Omowaye, B.I.O., Rastogi, N.K., Angersbach, A. and Knorr, D. (2003) Combined effects of pulsed electric field pre-treatment and partial osmotic dehydration on air drying behaviour of red bell pepper. *Journal of Food Engineering*, **60**, 89–98.

Aganovic, K., Siemer, C., Toepfl, S. and Heinz, V. (2012) Microscopic analysis of *Saccharomyces cerevisiae* and *Listeria innocua* after PEF, HPP and temperature treatment in Ringer's solution. Kongress Lebensmitteltechnologie GDL, Dresden.

Aguilar-Rosas, S F., Ballinas-Casarrubias, M.L., Nevarez-Moorillon, G.V., Martín-Belloso, O. and Ortega-Rivas, E. (2007) Thermal and pulsed electric fields pasteurization of apple juice: effects on physicochemical properties and flavour compounds. *Journal of Food Engineering*, **83**, 41–46.

Aguiló-Aguayo, I., Sobrino-López, Á., Soliva-Fortuny, R. and Martín-Belloso, O. (2008) Influence of high-intensity pulsed electric field processing on lipoxygenase and [beta]-glucosidase activities in strawberry juice. *Innovative Food Science and Emerging Technologies*, **9**, 455–462.

Aguiló-Aguayo, I., Oms-Oliu, G., Soliva-Fortuny, R. and Martín-Belloso, O. (2009) Changes in quality attributes throughout storage of strawberry juice processed by high-intensity pulsed electric fields or heat treatments. *LWT - Food Science and Technology*, **42**, 813–818.

Akdemir Evrendilek, G., Jin, Z.T., Ruhlman, K.T., Qiu, X., Zhang, Q.H. and Richter, E.R. (2000) Microbial safety and shelf-life of apple juice and cider processed by bench and pilot scale PEF systems. *Innovative Food Science and Emerging Technologies*, **1**, 77–86.

Álvarez, I., Raso, J., Palop, A. and Sala, F.J. (2000) Influence of different factors on the inactivation of *Salmonella senftenberg* by pulsed electric fields. *International Journal of Food Microbiology*, **55**, 143–146.

Amami, E., Vorobiev, E. and Kechaou, N. (2006) Modelling of mass transfer during osmotic dehydration of apple tissue pre-treated by pulsed electric field. *LWT - Food Science and Technology*, **39**, 1014–1021.

Amami, E., Fersi, A., Khezami, L., Vorobiev E. and Kechaou N. (2007a) Centrifugal osmotic dehydration and rehydration of carrot tissue pre-treated by pulsed electric field. *LWT - Food Science and Technology*, **40**, 1156–1166.

Amami, E., Fersi, A., Vorobiev, E. and Kechaou, N. (2007b) Osmotic dehydration of carrot tissue enhanced by pulsed electric field, salt and centrifugal force. *Journal of Food Engineering*, **83**, 605–613.

Angersbach, A. and Heinz, V. (1997) Elektrische Leitfähigkeit als Maß des Zellaufschlussgrades von zellulären Materialien durch Verarbeitungsprozesse. *Lebensmittelverfahrenstechnik*, **42**, 195–200.

Angersbach, A., Heinz, V. and Knorr, D. (2000) Effects of pulsed electric fields on cell membranes in real food systems. *Innovative Food Science and Emerging Technologies*, **1**, 135–149.

Barba, F.J., Jäger, H., Meneses, N., Esteve, M.J., Frígola, A. and Knorr, D. (2012) Evaluation of quality changes of blueberry juice during refrigerated storage after high-pressure and pulsed electric fields processing. *Innovative Food Science and Emerging Technologies*, **14**, 18–24.

Barbosa-Cánovas, G. V., Pothakamury, U. R., Palou E. and Swanson, B. G. (1998) *Nonthermal Preservation of Foods*. Marcel Dekker, New York, USA.

Barbosa-Cánovas, G.V., Góngora-Nieto, M.M., Pothakamury, U.R. and Swanson B.G. (1999a) Biological principles for microbial inactivation in electric fields. In: *Preservation of Foods with Pulsed Electric Fields*. Academic Press, San Diego, California, USA, pp. 47–75.

Barbosa-Cánovas, G.V., Góngora-Nieto, M.M., Pothakamury, U.R. and Swanson, B.G. (1999b) PEF inactivation of vegetative cells, spores, and enzymes in foods. In: *Preservation of Foods with Pulsed Electric Fields*. Academic Press, San Diego, Calilfornia, USA, pp. 108–155.

Bazhal, M. and Vorobiev, E. (2000) Electrical treatment of apple cossettes for intensifying juice pressing. *Journal of the Science of Food and Agriculture*, **80**, 1668–1674.

Bazhal, M.I., Lebovka, N.I. and Vorobiev, E. (2001) Pulsed electric field treatment of apple tissue during compression for juice extraction. *Journal of Food Engineering*, **50**, 129–139.

Bermúdez-Aguirre, D., Dunne, C.P. and Barbosa-Cánovas, G.V. (2012) Effect of processing parameters on inactivation of *Bacillus cereus* spores in milk using pulsed electric fields. *International Dairy Journal*, **24**, 13–21.

Bouzrara, H. and Vorobiev, E. (2003) Solid/liquid expression of cellular materials enhanced by pulsed electric field. *Chemical Engineering and Processing: Process Intensification*, **42**, 249–257.

Bushnell, A.H., Dunn, J.E., Clark, R.W. and Pearlman, J.S. (1993) High pulsed voltage systems for extending the shelf life of pumpable food products. US Patent **5**, 235–905.

Caminiti, I.M., Palgan, I., Noci, F., Munoz, A., Whyte, P., Cronin, D.A., Morgan D.J. and Lyng, J. G. (2011) The effect of pulsed electric fields (PEF) in combination with high intensity light pulses (HILP) on *Escherichia coli* inactivation and quality attributes in apple juice. *Innovative Food Science and Emerging Technologies*, **12**, 118–123.

Castro, A.J., Barbosa-Cánovas, G.V. and Swanson, B.G. (1993) Microbial inactivation of foods by pulsed electric fields. *Journal of Food Processing and Preservation*, **17**, 47–73.

Chalermchat, Y., Malangone, L. and Dejmek, P. (2010) Electropermeabilization of apple tissue: effect of cell size, cell size distribution and cell orientation. *Biosystems Engineering*, **105**, 357–366.

Charles-Rodríguez, A.V., Nevárez-Moorillón, G.V., Zhang, Q.H. andOrtega-Rivas, E. (2007) Comparison of thermal processing and pulsed electric fields treatment in pasteurization of apple juice. *Food and Bioproducts Processing*, **85**, 93–97.

Chen, F., Zeng, L., Zhang, Y., Liao, X., Ge, Y., Hu, X. and Jiang, L. (2009) Degradation behaviour of methamidophos and chlorpyrifos in apple juice treated with pulsed electric fields. *Food Chemistry*, **112**, 956–961.

Cho, H.-Y., Yousef, A.E. and Sastry, S.K. (1999) Kinetics of inactivation of *Bacillus subtilis* spores by continuous or intermittent ohmic and conventional heating. *Biotechnology and Bioengineering*, **62**, 368–372.

Corrales, M., Toepfl, S., Butz, P., Knorr, D. and Tauscher, B (2008) Extraction of anthocyanins from grape by-products assisted by ultrasonics, high hydrostatic pressure or pulsed electric fields: a comparison. *Innovative Food Science and Emerging Technologies*, **9**, 85–91.

Cserhalmi, Z., Vidács, I., Beczner, J. and Czukor, B. (2002) Inactivation of *Saccharomyces cerevisiae* and *Bacillus cereus* by pulsed electric fields technology. *Innovative Food Science and Emerging Technologies*, **3**, 41–45.

Cserhalmi, Z., Sass-Kiss, À., Tóth-Markus, M. and Lechner, N. (2006) Study of pulsed electric field treated citrus juices. *Innovative Food Science and Emerging Technologies*, **7**, 49–54.

Delgado, A.E. and Sun, D.W. (2001) Heat and mass transfer models for predicting freezing processes – a review. *Journal of Food Engineering*, **47**, 157–174.

Dimitrov, D.S. (1984) Electric field-induced breakdown of lipid bilayers and cell membranes: a thin viscoelastic film model. *Journal of Membrane Biology*, **78**, 53–60.

Dobreva, A., Tintchev, F., Heinz, V., Schulz, H. and Toepfl, S. (2010) Effect of pulsed electric fields (PEF) on oil yield and quality during destillation of white oil-bearing rose (*Rosa alba* L.). *Zeitschrift für Arznei- und Gewürzpflanzen*, **15**, 127–131.

Doevenspeck, H. (1960) Verfahren und Vorrichtung zur Gewinnung der einzelnen Phasen aus dispersen Systemen. EP 0 0637 91 A1, Germany.

Doevenspeck, H. (1961) Influencing cells and cell walls by electrostatic impulses. *Fleischwirtschaft*, **13**, 968–987.

El-Belghiti, K. and Vorobiev, E. (2004) Mass transfer of sugar from beets enhanced by pulsed electric field. *Food and Bioprocess Technology*, **82**, 226–230.

Elez-Martínez, P. and Martín-Belloso, O. (2007) Effects of high intensity pulsed electric field processing conditions on vitamin C and antioxidant capacity of orange juice and gazpacho, a cold vegetable soup. *Food Chemistry*, **102**, 201–209.

Eshtiaghi, M.N. and Knorr, D. (2002) High electric field pulse pretreatment: potential for sugar beet processing. *Journal of Food Engineering*, **52**(3), 265–272.

Fincan, M., DeVito, F. and Dejmek, P. (2004) Pulsed electric field treatment for solid–liquid extraction of red beetroot pigment. *Journal of Food Engineering*, **64**, 381–388.

García, D., Gómez, N., Raso, J. and Pagán, R. (2005a) Bacterial resistance after pulsed electric fields depending on the treatment medium pH. *Innovative Food Science and Emerging Technologies*, **6**, 388–395.

García, D., Hassani, M., Mañas, P., Condón, S. and Pagán, R. (2005b) Inactivation of *Escherichia coli* O157:H7 during the storage under refrigeration of apple juice treated by pulsed electric fields. *Journal of Food Safety*, **25**, 30–42.

Garde-Cerdán, T., Arias-Gil, M., Marsellés-Fontanet, A.R., Ancín-Azpilicueta, C. and Martín-Belloso, O. (2007) Effects of thermal and non-thermal processing treatments on fatty acids and free amino acids of grape juice. *Food Control*, **18**, 473–479.

Gaudreau, M.P.J., Hawkey, T., Petry, J. and Kempkes, M. (2004) Design considerations for pulsed electric field processing. 2nd European Pulsed Power Symposium, Hamburg, Shaker, Aachen.

Glaser, R.W., Leikin, S.L., Chernomordik, L.V., Pastushenko V.F. and Sokirko, A.I. (1988) Reversible electrical breakdown of lipid bilayers: formation and evolution of pores. *Biochimica et Biophysica Acta (BBA) – Biomembranes*, **940**, 275–287.

Grimi, N., Mamouni, F., Lebovka, N., Vorobiev, E. and Vaxelaire, J. (2011) Impact of apple processing modes on extracted juice quality: pressing assisted by pulsed electric fields. *Journal of Food Engineering*, **103**, 52–61.

Guderjan, M. (2006) Untersuchungen zum Einfluss elektrischer Hochspannungsimpulse bei der Gewinnung pflanzlicher Oele. Prozesswissenschaften, Technischen Universiät Berlin, Berlin, Germany.

Guderjan, M., Elez-Martinez, P. and Knorr, D. (2007) Application of pulsed electric fields at oil yield and content of functional food ingredients at the production of rapeseed oil. *Innovative Food Science and Emerging Technologies*, **8**, 55–62.

Guderjan, M., Toepfl, S., Angersbach, A. and Knorr, D. (2005) Impact of pulsed electric field treatment on the recovery and quality of plant oils. *Journal of Food Engineering*, **67**, 281–287.

Gurtler, J.B., Rivera, R.B., Zhang, H.Q. and Geveke, D.J. (2010) Selection of surrogate bacteria in place of *E. coli* O157:H7 and *Salmonella Typhimurium* for pulsed electric field treatment of orange juice. *International Journal of Food Microbiology*, **139**, 1–8.

Gurtler, J.B., Bailey, R.B., Geveke, D.J. and Zhang, H.Q. (2011) Pulsed electric field inactivation of *E. coli* O157:H7 and non-pathogenic surrogate *E. coli* in strawberry juice as influenced by sodium benzoate, potassium sorbate, and citric acid. *Food Control*, **22**, 1689–1694.

Gusbeth, C., Frey, W., Volkmann, H., Schwartz, T. and Bluhm, H. (2009) Pulsed electric field treatment for bacteria reduction and its impact on hospital wastewater. *Chemosphere*, **75**, 228–233.

Hamilton, W.A. and Sale A.J.H. (1967) Effects of high electric fields on microorganisms: 2. Mechanism of action of the lethal effect. *Biochimica et Biophysica Acta (BBA) – General Subjects*, **148**, 789–800.

Hartyáni, P., Dalmadi, I., Cserhalmi, Z., Kántor, D.-B., Tóth-Markus, M. and Sass-Kiss, À. (2011) Physical-chemical and sensory properties of pulsed electric field and high hydrostatic pressure treated citrus juices. *Innovative Food Science and Emerging Technologies*, **12**, 255–260.

Heinz, V., Alvarez, I., Angersbach, A. and Knorr, D. (2001) Preservation of liquid foods by high intensity pulsed electric fields – basic concepts for process design. *Trends in Food Science and Technology*, **12**, 103–111.

Hodgins, A.M., Mittal, G.S. and Griffiths, M.W. (2002) Pasteurization of fresh orange juice using low-energy pulsed electrical field. *Journal of Food Science*, **67**, 2294–2299.

Jalté, M., Lanoisellé, J.L. Lebovka, N.I. and Vorobiev, E. (2009) Freezing of potato tissue pre-treated by pulsed electric fields. *LWT - Food Science and Technology*, **42**, 576–580.

Jia, M., Zhang, H.Q. and Min, D.B. (1999) Pulsed electric field processing effects on flavor compounds and microorganisms of orange juice. *Food Chemistry*, **65**, 445–451.

Jin, Z.T. and Zhang, Q.H. (1999) Pulsed electric field inactivation of microorganisms and preservation of quality of cranberry juice. *Journal of Food Processing and Preservation*, **23**, 481–497.

Kern, M. (2005) Bipolarer Marxgenerator mit Schalterturm, direkter Triggerung und Gaskonditionierung für industriellen Dauerbetrieb, German Patent DE102004001782 A1.

Kessler, H.G. (1996) *Lebensmittel- und Bioverfahrenstechnik-Molkereitechnologie*. A. Kessler Verlag, Munich, Germany.

Knorr, D., Geulen, M., Grahl, T. and Sitzmann, W. (1994) Food application of high electric field pulses. *Trends in Food Science and Technology*, **5**, 71–75.

Kuthi, A., Alde, R., Gundersen, M. and Neuber, A. (2003) Marx generator using pseudospark switches. 14th IEEE International Pulsed Power Conference, 2003. Digest of Technical Papers, PPC-2003.

Lebovka, N.I., Shynkaryk, N.V. and Vorobiev, E. (2007) Pulsed electric field enhanced drying of potato tissue. *Journal of Food Engineering*, **78**, 606–613.

Loeffler, M. (2006) Generation and application of high intensity pulsed electric fields. In: *Pulsed Electric Field Technology for the Food Industry. Fundamentals and Applications*, Raso, J. and Heinz, V. (eds). Springer**,** Berlin, Germany, pp. 27–72.

Loginova, K.V., Lebovka, N.I. and Vorobiev, E. (2011) Pulsed electric field assisted aqueous extraction of colorants from red beet. *Journal of Food Engineering*, **106**, 127–133.

López, N., Puértolas, E., Condón, S., Raso, J. and Alvarez, I. (2009) Enhancement of the extraction of betanine from red beetroot by pulsed electric fields. *Journal of Food Engineering*, **90**, 60–66.

Marquez, V.O., Mittal, G.S. and Griffiths, M.W. (1997) Destruction and inhibition of bacterial spores by high voltage pulsed electric field. *Journal of Food Science*, **62**, 399–409.

Marsellés-Fontanet, À.R., Puig, A., Olmos, P., Mínguez-Sanz, S. and Martín-Belloso, O. (2009) Optimising the inactivation of grape juice spoilage organisms by pulse electric fields. *International Journal of Food Microbiology*, **130**, 159–165.

McDonald, C.J., Lloyd, S.W., Vitale, M.A., Petersson, K. and Innings, F. (2000) Effects of pulsed electric fields on microorganisms in orange juice using electric field strengths of 30 and 50 kV/cm. *Journal of Food Science*, **65**, 984–989.

Min, S., Jin, Z.T., Min, S.K., Yeom, H. and Zhang, Q.H. (2003) Commercial-scale pulsed electric field processing of orange juice. *Journal of Food Science*, **68**, 1265–1271.

Molinari, P., Pilosof, A.M.R. and Jagus, R.J. (2004) Effect of growth phase and inoculum size on the inactivation of *Saccharomyces cerevisiae* in fruit juices, by pulsed electric fields. *Food Research International*, **37**, 793–798.

Morales-de la Peña, M., Salvia-Trujillo, L., Rojas-Graü, M A. and Martín-Belloso, O. (2011) Impact of high intensity pulsed electric field on antioxidant properties and quality parameters of a fruit juice–soymilk beverage in chilled storage. *LWT - Food Science and Technology*, **43**, 872–881.

Mosqueda-Melgar, J., Raybaudi-Massilia, R. M. and Martín-Belloso, O. (2008a) Non-thermal pasteurization of fruit juices by combining high-intensity pulsed electric fields with natural antimicrobials. *Innovative Food Science and Emerging Technologies*, **9**, 328–340.

Mosqueda-Melgar, J., Raybaudi-Massilia, R.M. and Martín-Belloso, O. (2008b) Combination of high-intensity pulsed electric fields with natural antimicrobials to inactivate pathogenic microorganisms and extend the shelf-life of melon and watermelon juices. *Food Microbiology*, **25**, 479–491.

Mosqueda-Melgar, J., Raybaudi-Massilia, R.M. and Martín-Belloso, O. (2011) Microbiological shelf life and sensory evaluation of fruit juices treated by high-intensity pulsed electric fields and antimicrobials. *Food and Bioproducts Processing*, **90**, 205–214

Nguyen, P. and Mittal, G.S. (2007) Inactivation of naturally occurring microorganisms in tomato juice using pulsed electric field (PEF) with and without antimicrobials. *Chemical Engineering and Processing*, **46**, 360–365.

Noci, F., Riener, J., Walkling-Ribeiro, M., Cronin, D.A., Morgan, D.J. and Lyng, J.G. (2008) Ultraviolet irradiation and pulsed electric fields (PEF) in a hurdle strategy for the preservation of fresh apple Juice. *Journal of Food Engineering*, **85**, 141–146.

Pagán, R., Esplugas, S., Góngora, M., Barbosa-Canovas, G.V. and Swanson, B.G. (1998) Inactivation of *Bacillus subtilis* spores using high intensity pulsed electric fields in combination with other food conservation technologies. *Food Science and Technology International*, **4**, 33–44.

Plaza, L., Sánchez-Moreno, C., De Ancos, B., Elez-Martínez, P., Martín-Belloso, O. and Cano, M.P. (2011) Carotenoid and flavanone content during refrigerated storage of orange juice processed by high-pressure, pulsed electric fields and low pasteurization. *LWT - Food Science and Technology*, **44**, 834–839.

Pliquett, U., Joshi, R.P., Sridhara, V. and Schoenbach, K.H. (2007) High electrical field effects on cell membranes. *Bioelectrochemistry*, **70**, 275–282.

Puértolas, E., Lopez, N., Condón, S., Álvarez, I. and Raso, J. (2010) Potential applications of PEF to improve red wine quality. *Trends in Food Science and Technology*, **21**, 247–255.

Puértolas, E., López, N., Condón, S., Raso J. and Álvarez, I. (2009) Pulsed electric fields inactivation of wine spoilage yeast and bacteria. *International Journal of Food Microbiology*, **130**, 49–55.

Rastogi, N.K., Eshtiagi, M.N. and Knorr, D. (1999) Accelerated mass transfer during osmotic dehydration of high intensity electrical field pulse pretreated carrots. *Journal of Food Science*, **64**, 1020–1023.

Ravishankar, S., Zhang, H. and Kempkes, M.L. (2008) Pulsed electric fields. *Food Science and Technology International*, **14**, 429–432.

Riener, J., Noci, F., Cronin, D.A., Morgan, D.J. and Lyng, J.G. (2008) Combined effect of temperature and pulsed electric fields on apple juice peroxidase and polyphenoloxidase inactivation. *Food Chemistry*, **109**, 402–407.

Saldaña, G. (2011) Process criterion for the pasteurisation of food by pulsed electric fields. PhD Dissertation, Department of Animal Production and Food Science, University of Zaragoza, Spain.

Sánchez-Moreno, C., Plaza, L., Elez-Martínez, P., De Ancos, B., Martín-Belloso, O. and Cano, M.P. (2005) Impact of high pressure and pulsed electric fields on bioactive compounds and antioxidant activity of orange juice in comparison with traditional thermal processing. *Journal of Agricultural and Food Chemistry*, **53**, 4403–4409.

Schilling, S., Alber, T., Toepfl, S., Neidhart, S., Knorr, D., Schieber, A. and Carle, R. (2007) Effects of pulsed electric field treatment of apple mash on juice yield and quality attributes of apple juices. *Innovative Food Science and Emerging Technologies*, **8**, 127–134.

Schultheiss, C., Bluhm, H.J., Sack, M. and Kern, M. (2004) Principle of electroporation and development of industrial devices. *Zuckerindustrie*, **129**, 40–44.

Setlow, B., Atluri, S., Kitchel, R., Koziol-Dube, K. and Setlow, P. (2006) Role of dipicolonic acid in resistance and stability of spores of *Bacillus subtilis* with or without DNA-protective alpha/beta-type small acid-soluble proteins. *Journal of Bacteriology*, **188**, 3740–3747.

Sharma, S.K., Mulvaney, S.J. and Rizvi, S.S.H. (2000) *Food Process Engineering – Theory and Laboratory Experiments*. Wiley-Blackwell, New York, USA.

Shin, J.K., Lee, S.J., Cho, H.Y., Pyun, Y.R., Lee, J.H. and Chung, M.S. (2008) Germination and subsequent inactivation of *Bacillus subtilis* spores by pulsed electric field treatment. *Journal of Food Processing and Preservation*, **34**, 43–54.

Siemer, C., Elbers, K., Toepfl, S. and Heinz, V. (2011) Influence of pH value and conductivity on the inactivation of *B. subtilis* spores by PEF in combination with high temperature. Food Conference, Osnabrueck, Germany.

Siemer, C., Schmidgall, J., Toepfl, S. and Heinz, V. (2012) Chemical and functional properties of carrot mark andrind after HP and PEF treatment in comparison to a thermal treatment. Proceedings on Food Factory, Laval, France.

Siemer, C., Toepfl, S. and Heinz, V. (2014) Inactivation of B. subtilis spores by pulsed electric fields (PEF) in combination with thermal energy - I. Influence of process and product parameters. *Food Control* **39**, 163–171.

Sitzmann, W. (2006) *Technologieentwicklung der Elektroporation*. Reinbek, Germany.

Sitzmann, W. and Münch, E.W. (1988) Das ELCRACK ® Verfahren: Ein neues Verfahren zur Verarbeitung tierischer Rohstoffe. *Die Fleischmehlindustrie*, **40**(2), 22–28.

Spilimbergo, S., Dehghani, F., Bertucco A. and Foster, N.R. (2002) Inactivation of bacteria and spores by pulse electric field and high pressure CO_2 at low temperature. *Biotechnology and Bioengineering*, **82**, 118–125.

Taiwo, K.A., Eshtiaghi, M.N., Ade-Omowaye, B.I.O. and Knorr, D. (2003) Osmotic dehydration of strawberry halves: influence of osmotic agents and pretreatment methods on mass transfer and product characteristics. *International Journal of Food Science and Technology*, **38**, 693–707.

Toepfl, S. (2006) Pulsed electric fields (PEF) for permeabilization of cell membranes in food- and bioprocessing – applications, process and equipment design and cost analysis. PhD Thesis, Fakultät III, Prozesswissenschaften der Technischen Universität Berlin, Berlin, Germany.

Toepfl, S. (2011) Pulsed electric field food treatment – scale up from lab to industrial scale. *Procedia Food Science*, **1**, 776–779.

Toepfl, S. and Heinz, V. (2011) Pulsed electric field assisted extraction – a case study. In: *Nonthermal Processing Technologies for Food*, H.Q. Zhang, G.V. Barbosa-Cánovas, V.M. Balasubramaniam, C.P. Dunne, D.F. Darkas and J.T.C. Yuan (eds). Blackwell Publishing Ltd, Oxford, England, pp. 190–200.

Toepfl, S., Heinz, V. and Knorr, D. (2006) Applications of pulsed electric field technology for the food industry. In: *Pulsed Electric Field Technology for the Food Industry*, J. Raso and V. Heinz (eds). Springer, Berlin, Germany, pp. 197–221.

Toepfl, S., Heinz, V. and Knorr, D. (2007) High intensity pulsed electric fields applied for food preservation. *Chemical Engineering and Processing*, **46**, 537–546.

Torreggiani, D. (1993) Osmotic dehydration in fruit and vegetable processing. *Food Research International*, **26**, 59–68.

Torregrosa, F., Esteve, M.J., Frígola, A. and Cortésw, C. (2006) Ascorbic acid stability during refrigerated storage of orange-carrot juice treated by high pulsed electric field and comparison with pasteurized juice. *Journal of Food Engineering*, **73**, 339–345.

Tsong, T.Y. (1991) Electroporation of cell membranes. *Biophysical Journal*, **60**, 297–306.

Uemura, K. and Isobe, S. (2003) Developing a new apparatus for inactivating *Bacillus subtilis* spore in orange juice with a high electric field AC under pressurized conditions. *Journal of Food Engineering*, **56**, 325–329.

Walkling-Ribeiro, M., Noci, F., Cronin, D.A., Riener, J., Lyng J.G. and Morgan, D.J. (2008) Reduction of *Staphylococcus aureus* and quality changes in apple juice processed by ultraviolet irradiation, pre-heating and pulsed electric fields. *Journal of Food Engineering*, **89**, 267–273.

Walkling-Ribeiro, M., Noci, F., Cronin, D.A., Lyng J.G. and Morgan, D.J. (2009a) Shelf life and sensory attributes of a fruit smoothie-type beverage processed with moderate heat and pulsed electric fields. *LWT - Food Science and Technology*, **43**, 1067–1073.

Walkling-Ribeiro, M., Noci, F., Cronin, D.A., Lyng J.G. and Morgan, D.J. (2009b) Shelf life and sensory evaluation of orange juice after exposure to thermosonication and pulsed electric fields. *Food and Bioproducts Processing*, **87**, 102–107.

Weaver, J.C. (2000) Electroporation of cells and tissues. *IEEE Transactions on Plasma Science*, **28**(1), 24–33.

Wilhelm, C., Winterhalter, M., Zimmermann, U.and Benz, R. (1993) Kinetics of pore size during irreversible electrical breakdown of lipid bilayer membranes. *Biophysical Journal*, **64**, 121–128.

Wu, Y., Mittal, G.S. and Griffiths, M.W. (2005) Effect of pulsed electric field on the inactivation of microorganisms in grape juices with and without antimicrobials. *Biosystems Engineering*, **90**, 1–7.

Yin, Y. and He, G. (2008) A fast high-intensity pulsed electric fields (PEF)-assisted extraction of dissoluble calcium from bone. *Separation and Purification Technology*, **61**, 148–152.

Yonemoto, Y., Yamaguchi, H., Okayama, K., Kimura, A. and Murata, K. (1992) Characterization of microbial system for degradation of bacterial endospores. *Journal of Fermentation and Bioengineering*, **73**, 94–99.

Zhang, Q., Barbosa-Cánovas, G.V. and Swanson, B.G. (1995) Engineering aspects of pulsed electric field pasteurization. *Joumal of Food Engineering*, **25**, 261–281.

Zhang, Y., Hou, Y., Zhang, Y., Chen, J., Chen, F. Liao, X. and Hu, X. (2012) Reduction of diazinon and dimethoate in apple juice by pulsed electric field treatment. *Journal of the Science of Food and Agriculture*, **92**, 743–750.

Zimmermann, U., Pilwat, G., Beckers, F. and Riemann, F. (1976) Effects of external electrical fields on cell membranes. *Bioelectrochemistry and Bioenergetics*, **3**, 58–83.

27
Ohmic Heating

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27.1 Introduction

Ohmic heating (OH), also referred to as Joule heating, electrical resistance heating, direct electrical resistance heating, electroheating, and electroconductive heating, is defined as a process wherein (primarily alternating) electric currents are passed through foods or other materials with the primary purpose of heating them (Sastry and Barach, 2000). The concept of OH is not new and various attempts have been made to use it in food processing (Vicente, Castro and Teixeira, 2005; Ruan *et al.*, 2000). The first application of OH in food processing was started in the nineteenth century for milk pasteurization. It was reported that by 1938 this pasteurization technology was used in about 50 milk pasteurizers in five US states and served about 50 000 consumers. However, OH technology was not further developed in succeeding years due to the high processing cost and the lack of suitable inert electrode materials, except for the application of the electroconductive thawing process. Since the 1980s interest in OH has greatly recovered in the worldwide food industry for the following two reasons. First, high-temperature and short-time based aseptic processing technology has been considerably developed in the food industry but it has limitations to extend this technology for processing foods containing particulates. Ohmic heating as one of the alternative thermal processing technologies shows some specific advantages for a continuous high-temperature short-time process when used for foods containing large particulates. Second, with application of high-frequency power (up to 25–30 kHz), the electrodes, one of key components of the Ohmic heating system, can generally be made of stainless steel instead of titanium, which is required to avoid the chemical reaction between food and electrodes during the Ohmic heating process

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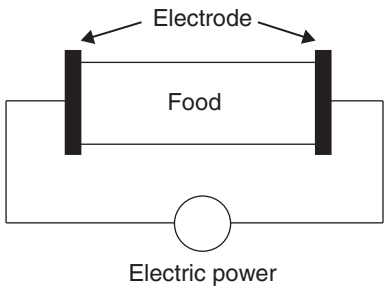


Figure 27.1 Diagram of Ohmic heating

if low-frequency power (60 Hz) is used. This has opened a wider door for the Ohmic heating technique to be implemented for industrial purposes (Chen, Abdelrahim and Beckerich, 2010).

The principle of OH is simple. As shown in Figure 27.1, in Ohmic heating, foods are made part of an electric circuit and the heat is directly generated inside foods when the electric power is applied and the current passes though them. The heating rate of foods is directly dependent on the electric voltage gradient and electric conductivity of foods. For the OH processing system used for commercial purposes, the electrical voltage gradient is automatically controlled by adjusting the power (voltage) to ensure the process can deliver the set process temperature at the end of heating. The electronic conductivity of the product, the most important processing factor, is varied from food to food and is also temperature dependent. If the product has more than one phase, such as in the case of a mixture of liquid and particulates, the electrical conductivity of all phases has to be considered. A difference in the electrical resistance and its temperature dependence between the two phases can make the heating characteristics of the system very complicated (Chen, Abdelrahim and Beckerich, 2010; Ruan *et al.*, 2000).

Compared with conventional heating methods, Ohmic heating has apparent advantages in terms of heating speed, uniformly heating, energy efficiency, low risk of fouling, and reducing maintenance cost. In addition, when OH is applied for continuous aseptic processing of foods containing particulates, it is possible to achieve the following potential advantages if the product formula and process are well designed:

1. A higher temperature in particulates than in liquid can be achieved, which is impossible for a conventional heating method based on aseptic processing.
2. The particle concentration can be high, up to 65% of the solid to total product.
3. The particle size can be large, up to 1 inch.

Therefore, aseptic processing of foods containing large particulates has been considered the most promising application of OH in the food industry (Knirsch *et al.*, 2002; Planiappan and Sastry, 2002; Wang *et al.*, 2001; Rice, 1995).

27.2 Applications of OH system

OH technology has been widely investigated and applied in a variety of food processing areas including blanching, pasteurization, sterilization, and extraction of food products (Icier, Yildiz and Baysal, 2005; Lima and Sastry, 1999; Mizrahi, 1996). Food products processed using OH can be solid, liquid, or a mixture of liquid and particulates. The early applications of OH were focused on processing of meat products, such as the cooking process of sausages and ham, and the thawing process of frozen meat products. The studies carried out by Fuchigami *et al.* (1994) and Ohtsuki (1991, 1993) indicated that OH technology can provide a uniform and quicker thawing process and can achieve a much better product quality in terms of colour and pH change. Piette *et al.* (2004) found that the cooking process time can be extremely reduced when OH was used for cooking brine-cured meat products (e.g. a ham weighing 1 kg was cooked in less than 2 min). However, attempts to develop a prototype version for a continuous cooking process were unsuccessful. Meat samples commonly have heterogeneous structures, which affects the uniform distribution of heat (Yildiz-Turp *et al.*, 2013). Compounds with poor conductivity, especially the fat in meat products, do not generate heat at the same rate as muscle, thus creating cold spots.

Since the 1980s, numerous studies of OH technology had been addressed on applications of aseptic processing for both liquid and liquid containing particulates. In the United States, a consortium of 25 partners from industry (food processors, equipment manufacturers, and ingredient suppliers), academia (food science, engineering, microbiology, and economics), and government was formed in 1992 to address practical challenges for OH to be applied for commercial purposes. A 5 kW pilot-scale continuous Ohmic heating system developed by APV Baker Ltd, Crawley, United Kingdom, was applied for investigation of a wide variety of low and high acid foods including broccoli and cheese, shrimp gumbo, strawberries in glaze, oriental chicken, and pasta primavera (shrimp, surimi, carrots, green beans, and mushrooms in alfredo sauce). These products were found to have equal or higher texture, color, and nutrient retention than those processed by traditional methods, such as freezing, retorting, and aseptic processing. In addition, an economic study was initiated. Ohmic operational costs were found to be comparable to those for freezing and retorting of low-acid products. Therefore, the consortium concluded that the OH technology was technically and economically viable.

Many published research studies using pilot or commercial continuous Ohmic production units were led with the collaboration of the US Army Natrick Research Development and Engineering Center (NRDEC). This organization was interested in the potentiality of this process to provide rations of excellent quality, with a long shelf-life at ambient temperature. The research project conducted by Kim *et al.* (1996) was focused on the lethality within food particles undergoing Ohmic heating using microbiological and chemical marker measurements. The methodology developed was then used to validate the process in a 25 kW Ohmic heating system for a food containing 35% of particles and 65% of starch solution.

In 2005, Campbell Soup Company (Camden, New Jersey, USA) built up an Ohmic aseptic production line in one of the Campbell plants, located at Le Pontet, France, producing different types of soups, low-acid foods containing large particulates of vegetables and/or meats. The Ohmic aseptic processing system is manufactured by Rossi & Catelli, Italy. The process is developed using a computer prediction model validated by biological tests. In 2010, the US Food and Drug Administration (FDA) accepted the first filing of Ohmic heating process used for low-acid food containing vegetable particulates with a size of $^3/_4$ inch from Campbell Soup Company.

27.3 OH heating process and equipment

27.3.1 Process flow

The OH process can be designed as a batch system or continuous system. Figure 27.2 shows a typical configuration for a batch Ohmic heating process. The heating cell consists of a cylinder box and two electrodes located at each extremity. The continuous OH process can be designed in different types, depending on the manufacturer (Vicenti, Castro and Teixeira, 2005). It can be a simple tube with pairs of opposing electrodes mounted on the tube walls opposite to each other (Figure 27.2a), coaxial tubes acting as electrodes with the food flowing between them (Figure 27.2b), or a vertical tube with the electrodes embodied at regular intervals (Figure 27.2c). Since the electric field is perpendicular to the food flow for the equipment represented in Figure 27.2b, this configuration is often called cross-field. As the electric field is parallel to the food flow (Figure 27.2b and 27.2c), the configuration is termed in-field. In the cross-field configuration the electric field strength is constant. For the in-field configuration, the electric conductivity will increase and therefore the field strength experienced by the product will increase as it approaches the outlet because the product will heat during its path through the heater. To minimize this effect, when multiple electrodes are used in series (Figure 27.2c), they are spaced differently to account for the increase of electric conductivity with temperature. As shown in Figure 27.2c, the second pair of electrodes has a bigger space than the first one.

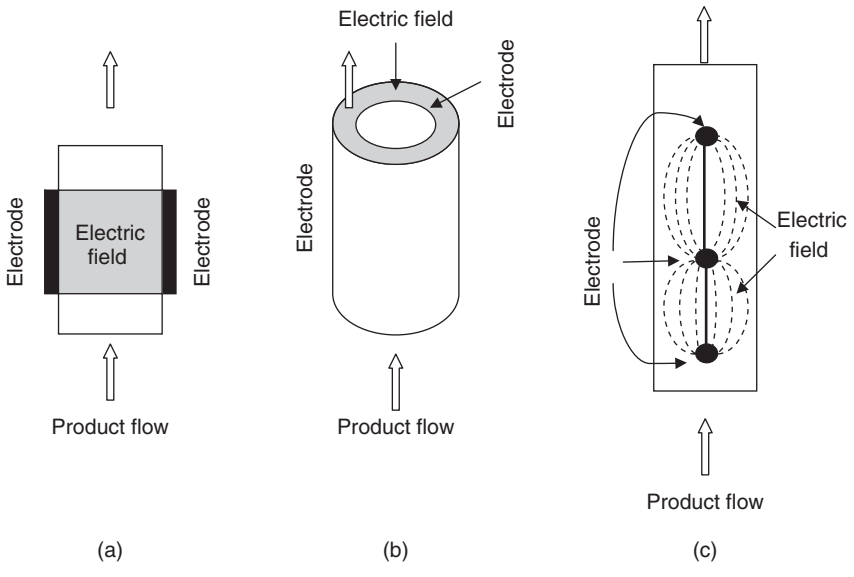


Figure 27.2 Types of Ohmic heating processes

Selection of the OH process is dependent on the foods being processed and objectives of the process, such as blanching, cooking, pasteurization, and sterilization. Usually, the batch OH process is applied for cooking solid foods such as meat products, while the thawing process, a continuous process, is mostly used for liquid foods or mixture of liquid and particulates.

27.3.2 Commercial equipment

The first continuous OH aseptic process was designed by the United Kingdom Electricity Research and Development Center and was licensed to APV Baker Ltd for commercial application in 1983 (Eliot-Godereaux, Zuber and Goullieux, 2001; Skudder, 1992). Figure 27.3 shows the diagram of a 10 kW continuous Ohmic heating system made by APV. It consists of a vertical heating column, holding and cooling tubes. The heating section contains three housings, each having a cantilever electrode. The electrode housings are connected by spacer tubes giving two heating sections, as shown in Figure 27.3. A piston pump is used for introducing the product to the system, which is designed to allow high viscous food and particles to pass through with minimal damage. The system power is 10 kW, allowing a 75 °C temperature increase with a flow rate of 100 kg/h.

Figure 27.4 is a diagram of the commercial OH heating processing system from Raztek, a company located in the United States. It uses common three-phase high-voltage AC power available worldwide with a frequency of

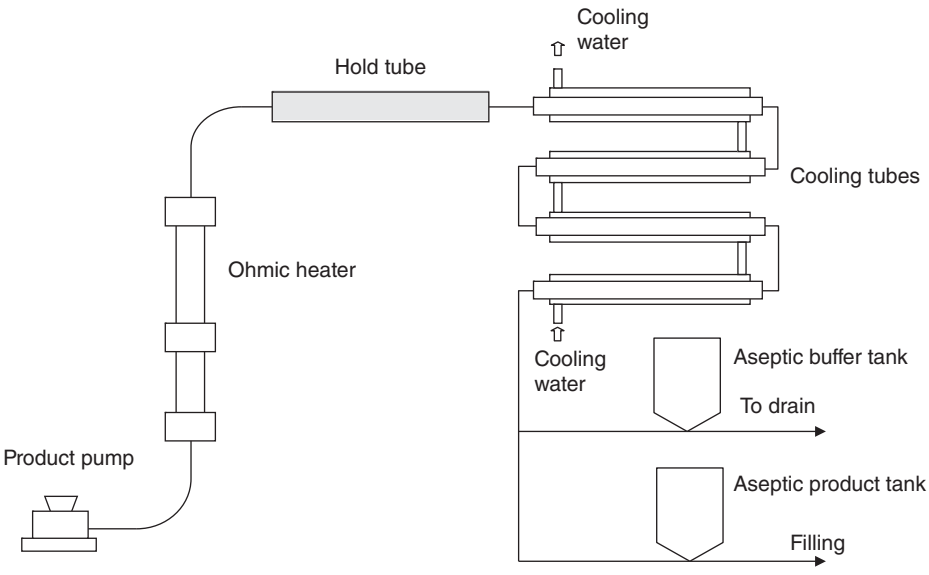


Figure 27.3 Diagram of a 10 kW continuous Ohmic heating system made by APV (source: Eliot-Godereaux *et al.*, 2001. Reproduced with permission of Elsevier)

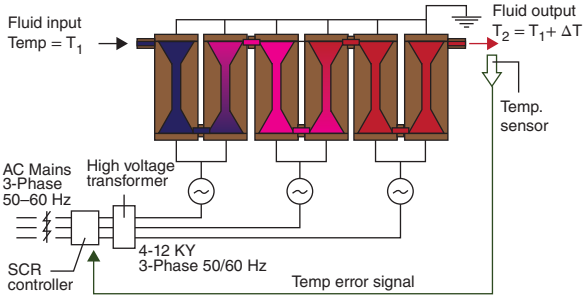


Figure 27.4 Commercial Ohmic heating processing system. Source: presentation by David Reznik for the Aseptipak Global Forum, 2001, Chicago, USA. Image reproduced with kind permission from Raztek, California, USA. *See plate section for colour version*

50 or 60 Hz. The electrodes are made of specially designed graphite electrodes to avoid metal dissolution by electrolysis.

Figure 27.5 shows an Ohmic aseptic processing system. The whole process consists of three sections: heating, holding, and cooling. The heating section consists of two Ohmic heaters, between which there is an intermediate holding tube to equalize the temperature difference between different phases during the heating process. Each Ohmic heater has four heating tubes, as

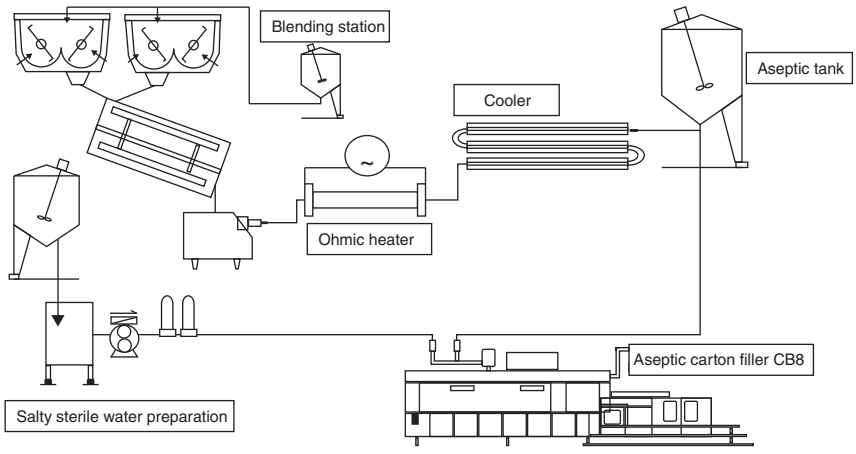


Figure 27.5 Ohmic aseptic processing system

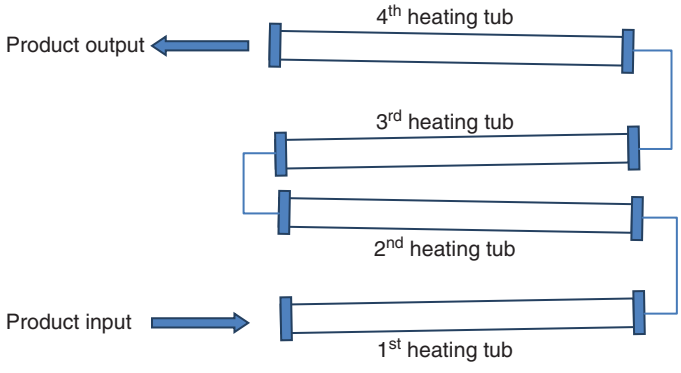


Figure 27.6 Diagram of a heating cell

shown in Figure 27.6. The total power used for both heaters is 240 kW, with an adjustable voltage up to 2000 V.

27.4 Modelling of the OH process

Computer modeling has become a useful and necessary tool for development of various thermal processes, especially for aseptic processing of foods containing particulates, where internal temperatures cannot be measured without significantly interfering with the product flow. The problem is more

acute during Ohmic heating because of the presence of an electrical field (Sastry and Li, 1996). Two modeling methods have been developed and applied for simulating Ohmic heating by Sastry and Palaniappan (1992) and De Alwis and Fryer (1990).

The model developed by De Alwis and Fryer (1990) was based on the solution to Laplace's equation to calculate the heat generation rate together with the transient energy balance equation to model a single particle immersed in a fluid medium without convection. This model has been extended by Zhang and Fryer (1990) to include multiple spheres uniformly distributed on a lattice within a nonconvective fluid.

The electric field or voltage distribution can be developed from Maxwell's equations or by combining Ohm's law and the continuity equation for an electrical current:

$$\nabla(\sigma_i \nabla V) = 0 \tag{27.1}$$

where V = voltage, ∇ = gradient, and σ_i = electrical conductivity of phase i, which can take different values for the particles and liquid.

Ignoring convection effects, the heat transfer problem is one of pure conduction with internal energy generation:

$$\rho_i C_{pi} \frac{\partial T}{\partial t} = \nabla(k_i \nabla T) + \dot{u}_i \tag{27.2}$$

where i again represents the phase, k is thermal conductivity, $\dot{u}$ is specific internal energy generation rate, ρ is density, C_p is specific heat capacity, T is temperature, and t is time.

The external boundary condition is one of convection to the surroundings:

$$-k_{iS} \nabla T \boldsymbol{n} = U(T_{iS} - T_\infty) \tag{27.3}$$

where k_{iS} is thermal conductivity of phase i at the surface, n is unit normal vector, U is overall heat transfer coefficient, T_{iS} is surface temperature of phase i, and T_∞ is surrounding temperature.

The internal energy generation term in Equation (27.3) is given by

$$\dot{u}_i = |\nabla V|^2 \sigma_{0i}(1 + m_i T) \tag{27.4}$$

where ∇V is voltage gradient, σ_{0i} is initial electrical conductivity, m_i is temperature conpensation constant, and T is temperature.

The system of equations (27.1) to (27.4) can be solved by the Galerkin-Crank-Nicolson algorithm, a hybrid spatially finite element, temporally a finite difference scheme.

Another modeling method developed by Sastry and Palaniappan (1992) using the electric circuit analogy has been applied for the simulation of continuous processing of foods containing particulates. For a continuous process

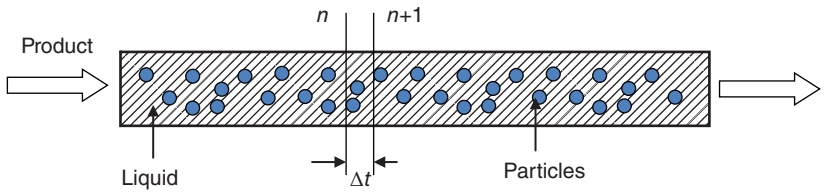


Figure 27.7 Continuous process for processing of multiphase foods

used for processing multiphase foods, as shown in Figure 27.7, the energy balance equation can be described by the equation

$$\dot{v}_f \rho_f C_{pf}(T_f^{n+1} - T_f^n) = \sigma_f E^2 v_f + N_p h_{fp} A_p (T_{sm} - T_f^n) + U A_{pi}(T_a - T_f^n) \quad (27.5)$$

where v is the volume flow rate, ρ is the density, v is the volume, C_p is the heat capacity, T is the temperature, E is the electric field strength, σ is the electric conductivity, n is the incremental section number in the pipe length, N is the number of particulates, h is the heat transfer coefficient between the carrier fluid and the particle surface, A is the surface area, and U is the overall heat transfer coefficient. Subscripts *f, p, pi, a* and *sm* represent carrier fluid, particle, pipe, air and surface mean, respectively; superscript n is the location or time step.

The heat penetration for particles is the conduction heating equation with temperature dependent internal energy generation.

$$\rho_p C_{pp} \frac{\partial T}{\partial t} = \nabla \cdot (k_p \nabla T_p) + \sigma_p E^2 \quad (27.6)$$

where subscript p represents the particle and k is the thermal conductivity.

With a time-dependent boundary condition

$$k_p \nabla T_p \cdot \boldsymbol{n} = h_{fp}(T_{sp} - T_f) \quad (27.7)$$

where T_{sp} is the particle surface temperature. From the equations (27.5) and (27.6), it can be easily seen that the internal heat generated by electricity is dependent on two factors: conductivity (σ) and electrical field strength (E). The electrical conductivity and its temperature dependency for different materials can be measured and described by the following linear equation:

$$\sigma = \sigma_0 + m(T - T_0) \quad \text{or} \quad \sigma = \sigma_{70} + m(T - 70) \quad (27.8)$$

where subscript 0 represents a reference temperatureand m is the temperature coefficient.

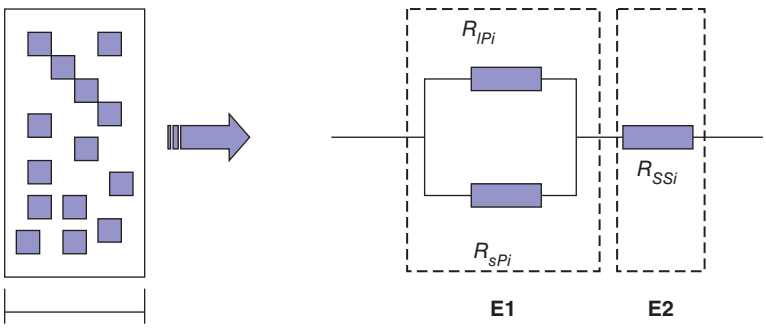


Figure 27.8 A set of equivalent resistances in series

The electrical field distribution was solved by use of the method developed by Sastry and Palaniappan (1992) via circuit theory, in which the Ohmic heater column is considered to be a set of equivalent resistances in series (Figure 27.8). The resistances in each increment section are calculated using a continuous (carrier fluid) and discontinuous (solid) phase as

$$R_i = R_{lSi} + \frac{R_{lPi}R_{sPi}}{R_{lPi} + R_{sPi}} \tag{27.9}$$

where R_{lSi} is the resistance of carrier fluid in series, R_{lPi} is the resistance of carrier fluid in parallel, and R_{sPi} is the resistance of particles in parallel. The total resistance of the Ohmic heater column is then

$$R = \sum_{n=1}^{N} R_i \tag{27.10}$$

The total current flowing through the system is

$$I = \frac{\Delta V}{R} \tag{27.11}$$

The voltage drop over increment I was calculated as

$$\Delta V_i = IR_i \tag{27.12}$$

and the results were used to calculate the voltage gradient and energy generation within each incremental section.

This modelling method has been applied for developing a comprehensive computer modeling program as the process calculation tool in commercial aseptic products using OH (Chen, Abdelrahim and Beckerich, 2010; Salengke and Sastry, 2007).

27.5 Critical factors of OH processing

27.5.1 Effects of electrical conductivities

Ohmic heating rates are critically dependent on the electrical conductivity of the food being processed. Key considerations in the electrical conductivity include overall conductivity, the difference between carrier fluid and particles, and nonconductive materials.

De Alwis, Halden and Fryer (1989) provided details on the effects of electrical conductivity on the Ohmic heating process. For a given Ohmic heating system, in order to achieve the desired heat rate (normally in the range of 1–5 °C/s), the amount of electrical power (P) required can be estimated using the product flow rate (m), specific heat capacity (C_p), and density (ρ), together with the inlet and outlet temperatures:

$$P = m\rho C_p(T_{out} - T_{in}) \tag{27.13}$$

The current (I) and voltage (V) required to deliver that power can then be found using standard electrical relationships:

$$I = \sqrt{\frac{P}{R}}$$
$$V = \sqrt{PR} \tag{27.14}$$

where R is the overall system resistance. There is a limitation for voltage and current that can be applied to a given commercial system. The maximum safe current will depend on the current density that the system can sustain, a function of both electrode and system geometry. The maximum voltage that can be sustained in a commercial system will be limited by the cost of electrical and control hardware.

These limitations will restrict the range of conductivities for which Ohmic heating is possible. In the limit of very low electrical conductivity the resistance of the food will be so high that it will be impossible to get a satisfactory heating rate using the highest possible voltage. In the limit of very high electrical conductivity, in contrast, the current will be over the limit that the system can sustain. Therefore, for a given Ohmic heating system, prior to processing, the conductivities of food materials should be tested to make sure that they are in the acceptable range.

For processing heterogeneous food mixtures of liquid and particles, the conductivity difference between the carrier fluid and particles is the key parameter affecting the heating behavior of both liquid and particles during Ohmic heating. A solid immersed in a liquid can either heat faster or slower than the liquid. This effect has been found to be a function of both solid and liquid

electrical conductivity and the orientation of the particles to the electric field (de Alwis, Halden and Fryer, 1989). If particles and liquid are connected in series type, then the higher conductivity part will heat slower. In other words, if their connection is parallel, then the higher conductivity will heat faster.

27.5.2 Effects of particle size and concentration

Various combinations of meats, vegetables, pasta, and fruits can be successfully processed when accompanied by an appropriate carrier medium and suitable process controls. Basically, there are three fundamental particulate considerations including size, shape, and concentration. Optimizing the correct combination of these characteristics will result in excellent texture through uniformity of particulate heating.

As mentioned previously, one of obvious advantages to using Ohmic heating is its capability to process large particulate foods, which are difficult for a conventional heating method. Technically, for the Ohmic heating process, the particulate size should be determined by the following concerns: pipe diameter and aseptic filler capability in the particulate size. In fact, the consumer response suggests that particulates larger than 1 in^3 would require cutting prior to consuming and thereby reduce convenience. Thus, the 1 in^3 is usually considered as the limit size for the Ohmic heating process (Zoltai and Swearingen, 1996). In addition, if particulates have a significantly lower electrical conductivity than the carrier medium, the former will be heated slower than the latter during Ohmic heating. As particulates increase in size, with the surface-to-volume ratios decreasing, the heat transfer between particulates and the carrier medium is affected.

For the continuous Ohmic heating process, the plug flow is the desired type as it can create a uniform residence time for products to pass through the pipes. In order to have this result, foods with a formulation of high concentration particulates are commonly used for the Ohmic heating process. Particulate concentration in most Ohmic formulations ranges from 20 to 70%. Extremely low or high concentrations require special consideration of size, shape, and texture to optimize stability of the formulation during processing. Higher concentrations can be processed if the particulates are pliable and small and their geometry is varied, as this decreases the voids between particles. Lower concentrations generally require a higher viscosity carrier medium to maintain particulate suspension.

27.5.3 Effects of carrier medium viscosity

The main function of the carrier fluid is to hold the particulates in a suspended state throughout the entire process. Usually, low carrier fluid viscosity can be used for products containing a high concentration of particulates, while

high carrier fluid viscosity should be used for those with a low particulate concentration. If the viscosity is inadequate, or if significant thinning occurs during processing, particulates may settle and/or carrier fluid may flow past the particulates without suspending them. This will result in substantial unevenness of heating. On the other hand, if the carrier fluid is highly viscous, particulate abrasion can cause an effect in textural integrity and significant system backpressure problems may also occur. Changes in viscosity during processing, such as starch gelatinization or release of moisture from particulates, can also result in unevenness of heating. Therefore, in practical applications, it is important that the starch solution should be pre-gelatinized to avoid a phase change during processing.

The effects of fluid viscosities on Ohmic heating rates of fluid–particle mixtures were investigated by Khalaf and Sastry (1996). Results indicated that fluids of identical electrical conductivity but different viscosity were used with identical amounts of solid particles (of electrical conductivity lower than the fluid) and heated ohmically in batch (static and vibrating) and continuous flow heaters. In the static Ohmic heater, the heating rate of the fluid and particles was found to be comparable for the different fluids. However, in the vibrating Ohmic heater, the heating rate of fluid and particles was found to increase with increasing fluid viscosity. In the continuous flow Ohmic heater, the mixture with the higher viscosity fluid heated faster than that with the lower viscosity fluid. The important implication is that poor interphase convective heat transfer may actually contribute to accelerated overall heating, since the more (electrically) conductive phase does not lose heat readily to the less (electrically) conductive phase, and consequently heats rapidly, transferring heat to the other phase by larger temperature differences.

27.6 Sensitivity analysis of the continuous OH system

The process sensitivity analysis is one of the important steps for a successful process development since it can provide basic knowledge of how the variation in the output of the process can be apportioned, qualitatively or quantitatively, to different sources of variation in the processing conditions. The process sensitivity analysis of a real continuous Ohmic heating system for soup products containing large particulates was performed by Chen, Abdelrahim and Beckerich (2010) using a validated computer modeling package. The major processing variables selected for the sensitivity analysis included conductivities of both particle (σ_p) and liquid (σ_f), surface heat transfer coefficient of particles, size of particle (S_p), particle concentration (P_c), flow rate (V_f), and initial temperature (T_i). The response variables used for the sensitivity analysis were: the carrier fluid temperature at the end of holding tube (T_L),

particle center temperatures (TP_1 and TP_2) at the entrance and end of the holding tube, and accumulated lethality values of the carrier fluid (FL), particle center (F_o), and particle integrated (F_v) at the end of the holding tube. The process temperature for the carrier fluid at the entrance of the holding tube was fixed at 133 °C. The results indicated that electrical conductivity of both the carrier fluid and particles are the most sensitive variables in the process temperature and target lethality values. Under a given control temperature at the end of heating, increasing carrier fluid conductivity would result in lower process temperatures for the carrier fluid and particles at the end of the holding tube, while increasing particle conductivity would generate higher process temperatures for them. It is possible for particles to achieve a higher temperature during the Ohmic heating process than the carrier fluid if the particle conductivity is increased close to the carrier fluid conductivity. Figures 27.9 and 27.10 illustrate the effects of conductivity of the carrier fluid and particles on the heating behaviors during OH heating processing.

Other processing variables such as particle size, particle concentration, flow rate, and initial product temperature also need to be controlled and monitored during the process in order to achieve the expected process temperature and lethality value at the end of the holding tube. Relatively, thermal diffusivity and the surface heat transfer coefficient between the carrier fluid and particles are weak effects on the process temperature and accumulated lethality value for both the carrier fluid and particles within the ranges investigated.

27.7 Conclusions

OH, as an alternative emerging heating method, has been widely applied in food processing areas including blanching, pre-heating, sterilization and thawing processes. Specifically, with development of the high-frequency power system and online processing control technology in terms of meeting food safety requirements, OH technology has become one of the alternative thermal process technologies to update the traditional aseptic processing system, which makes it possible to extend the high-temperature short-time (HTST) application from liquid foods to particulate foods. It has been demonstrated that the OH process can provide unique advantages over traditional heating methods using various heat exchangers, such as rapid and uniform heating, high-quality products, less fouling and greater energy efficiency. However, it should be noted that OH technology, like most of the other emerging processing technologies, has also some disadvantages or limitations for its application. First of all, the application of OH is limited to products with ionic content (i.e. it is not suitable for oils, etc.). In order to achieve uniform temperature, the difference

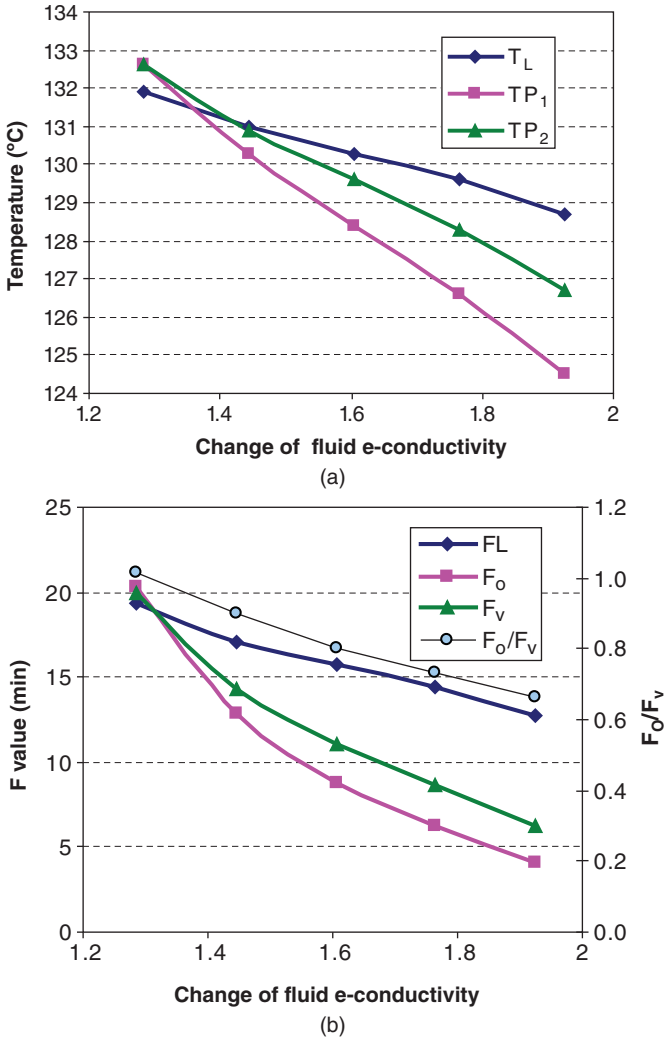
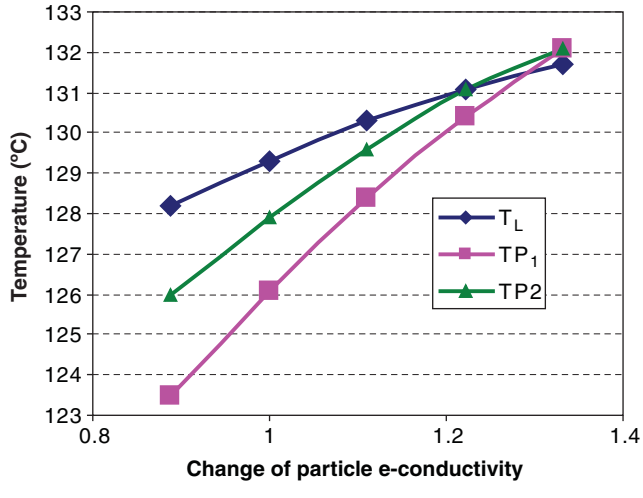
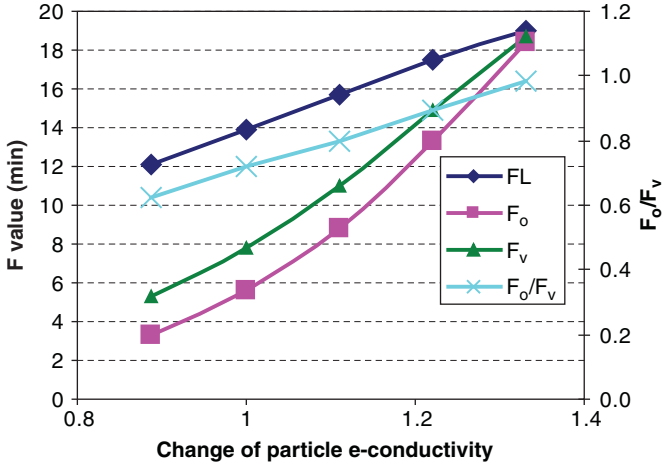


Figure 27.9 Effects of liquid conductivity (σ_f) on process temperatures and lethality values of both liquid and particles such as (a) effects of fluid e-conductivity on temperatures, and (b) effects of fluid e-conductivity on F values

of the electric property between ingredients must be controlled and it needs a very high level control system. Finally, it is challenging to directly measure the particle temperature during a continuous OH heating process; thus the process developed for multiphase products must be validated biologically.



(a)



(b)

Figure 27.10 Effects of particle conductivity (σ_p) on process temperatures and lethality values of both liquid and particles such as (a) effects of particle e-conductivity on temperatures, and (b) effects of particle electrical conductivity on F values

References

Chen, C.R., Abdelrahim, K. and Beckerich, I. (2010) Sensitivity analysis of continuous Ohmic heating process for multiphase foods. *Journal of Food Engineering*, **98**(2), 257–265.

de Alwis, A.A.P. and Fryer, P.J. (1990) A finite element analysis of heat generation and transfer during Ohmic heating of food. *Chemical Engineering and Science*, **45**(6), 1547–1559.

de Alwis, A.A.P., Halden, K. and Fryer, P.J. (1989) Shape and conductivity effects in the Ohmic heating of foods. *Chemical Engineering Research and Design*, **67**, 159–168.

Eliot-Godereaux, S.C., Zuber, F. and Goullieux, A. (2001) Processing and stabilisation of cauliflower by Ohmic heating technology. *Innovative Food Science and Engineering Technologies*, **2**, 279–287.

Fuchigami, M., Hyakuumoto, N., Miyazaki, K., Nomura, T. and Sasaki, J. (1994) Texture and histological structure of carrots frozen at a programmed rate and thawed in an electrostatic field. *Journal of Food Science*, **59**(6), 1162–1167.

Icier, F., Yildiz, H. and Baysal, T. (2005) The effect of Ohmic heating enzyme activity. Proceedings of the 2nd Internal Conference of the Food Industries and Nutrition Division on Future Trends in Food Science and Nutrition, 27–29 November, Cairo, Egypt, 60 pp.

Khalaf, W.G. and Sastry, S.K. (1996) Effect of fluid viscosity on the Ohmic heating rate of solid–liquid mixtures. *Journal of Food Engineering*, **27**, 145–158.

Kim, H.J., Choi, Y.M., Yang, T.C.S., Taub, I.A., Tempest, P., Skudder, P., Tucker, G. and Parrot, D.L. (1996) Validation of OH for quality enhancement of food products. *Food Technology*, **50**, 253–261.

Knirsch, M.C., Santos, C.A., Vicente, A.A. and Penna, T.C. (2010) Ohmic heating – a review. *Trends in Food Science and Technoloy*, **21**, 436–441.

Lima, M. and Sastry, S.K. (1999) The effects of Ohmic heating frequency on hot-air drying rate and juice yield. *Journal of Food Engineering*, **41**, 115–119.

Mizrahi, S. (1996) Leaching of soluble solids during blanching of vegetables by Ohmic heating. *Journal of Food Engineering*, **29**, 153–166.

Ohtsuki, T. (1991) Process for thawing foodstuffs. US Patent **5**, 034–236.

Ohtsuki, T. (1993) Process for thawing foodstuffs. European Patent 0409430.

Piette, G., Buteau, M.L., de Halleux, D., Chiu, L. and Ramaswamy, H.S. (2004) Ohmic cooking of processed meats and its effects on product quality. *Journal of Food Science*, **69**, 71–78.

Planiappan, S. and Sastry, S. (2002) Ohmic heating. In: *Control of Foodborne Microorganisms*, Juneja, V.K. and Sofos, J.N. (eds). Marcel Dekker, New York, USA, pp. 451–460.

Rice, J. (1995) Ohmic adventures. *Food Processing*, **56**(3), 87–91.

Ruan, R., Ye, X., Chen, P. and Doona, C.J. (2000) Ohmic heating. In: *Thermal Technologies in Food Processing*, edited by Philip Richardson, P. (ed.). CRC Press, Boca Raton, Florida, USA, pp. 241–265.

Salengke, S. and Sastry, S.K. (2007) Experimental investigation of Ohmic heating of solid–liquid mixtures under worst-case heating scenarios. *Journal of Food Engineering*, **83**, 324–336.

Sastry, S.K. and Barach, J.T. (2000) Ohmic and inductive heating. *Journal of Food Science*, **65**, 42–46.

Sastry, S.K. and Li, Q. (1996) Modeling the Ohmic heating of foods. *Food Technology*, **50**(5), 246–248.

Sastry, S.K. and Palaniappan, S. (1992) Ohmic heating of liquid–particle mixtures. *Food Technology*, **46**(12), 64.

Skudder, P.J. (1992) Long life products by Ohmic heating. *International Food Ingredients*, **4**, 36–41.

Vicente, A.A., Castro,I. and Teixeira, J.A. (2005) Ohmic heating for food processing: new technologies and quality issues. In: *Thermal Processing in Food Processing*, Sun, D.W. (ed.). CRC Press, Boca Raton, Florida, USA, pp. 426–468.

Wang, C.S., Kuo, S.Z., Kuo-Huang, L.L. and Wu, J.S. (2001) Effect of tissue infrastructure on electric conductance of vegetable stems. *Journal of Food Science: Food Engineering and Physical Properties*, **66**(2), 284–288.

Yildiz-Turp, G., Sengun, I.Y., Kendirci, P. and Icier, F. (2013) Effect of Ohmic treatment on quality characteristic of meat: a review. *Meat Science*, **93**, 444–448.

Zhang, L. and Fryer, P.J. (1990) Models for the electrical heating of solid–liquid food mixtures. *Chemical Engineering Science*, **48**, 633–642.

Zoltai, P. and Swearingen, P. (1996) Product development considerations for Ohmic processing. *Food Technology*, **50**(5), 263–266.

28

Intelligent Identification System for Poultry Portion Sorting

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28.1 Introduction

Food processing plants have come a long way since all processing stages were carried out manually by human labourers. Nowadays, robots are either replacing workers or aiding them in performing most of the processing tasks. Of course, the need for human supervision throughout processing is yet to be eliminated, but it seems that we are heading that way. With the rapid advancement in technology, we foresee completely autonomous processing plants, that is human-free plants.

One may ask why we seek to replace human workers with mechanized robots. Here, we must consider a number of reasons, which vary according to different products. There could be technical, economical, health or classified aspects to consider. Regardless of which reason or what aspect, we seem to be heading towards that goal – that is robots taking over in processing plants.

While robots have been successfully performing complex and accurate operations in industry, they still lag behind humans when it comes to decision making. Human decision makers are more adaptive to changes that may occur during a process, which is simply due to our natural intelligence. Thus, for robotic systems to match this flexible human ability, they require intelligence – to be precise, artificial intelligence. The combination of artificially

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intelligent systems with robotic systems is what the future production and processing plants seek, and we are not far off that target.

So what is artificial intelligence (AI) all about? In simple terms, we may describe AI as the 'missing link' to make robots and systems autonomous and human-independent during their operations. AI is about mimicking the way we humans make decisions based upon received stimuli, which vary according to our input receivers. Human senses such as seeing, hearing, smelling, tasting and touching comprise the major input stimuli processes when making decisions. Luckily, few of these human natural processes can be easily modelled artificially in robotic systems; for example a camera for an eye, a microphone for an ear and so on. Therefore, we could provide systems with a 'seeing' capability, for example, and process the received information via image processing, but the challenge here is what to make out of the received information (or in this example the received visual data). That is where artificial intelligence comes in – to make sense of what a robotic system receives.

The field of artificial intelligence has been expanding rapidly over the past three decades, with different techniques often emerging offering intelligent solutions to different problems. However, the core of artificial intelligence still relies on artificial neural networks. This is so for a good reason; neural networks mimic or model the human brain's structure and function. The brain is naturally where perception (making sense of received information) and decisions are made.

In this chapter, we consider a food processing problem, in particular a poultry processing problem, and focus on a process that is often carried out by human workers, namely sorting raw poultry portions in a processing plant. Poultry meat in general and chicken meat in particular has become the world's second most consumed type of meat (Somsen, Capelli and Tramper, 2004), thus making poultry production a highly competitive industry throughout the world. Consequently, feed suppliers, producers, manufacturers of equipment, and all related businesses to poultry production continue to support research works that aim at providing faster automated processing for poultry production.

28.2 Automation in poultry processing

In modern poultry processing plants, the slaughter of birds and then cutting up the carcass is a fully automated process without the handling of human labourers (Gainco Inc., 2011a; Van Hoogen, 2005; Sams, 2001). This is important not only because it increases the speed of production but it also reduces the potential of contamination from human operators to the raw poultry carcasses and vice versa (Keener *et al.*, 2004).

The next process after cutting up the bird carcass in a poultry plant is to separate or sort the portions into different containers, followed by packaging.

Currently, few systems use an automated method based on the physical weight of a portion as an indicator for sorting (Gainco Inc., 2011b; Sams, 2001); however, manual sorting continues to be the most prevalent method used for sorting poultry portions prior to packaging. Inherently, there are problems with such manual sorting methods, including inconsistency, high labour costs, worker fatigue and increasing employment costs; these have all been identified as the important factors driving the demand for automation of the industry (Jarimopas and Jaisin, 2008). Moreover, we have become more cautious about handling raw meat and poultry products since the spread of diseases such as bird flu; therefore, finding an efficient method to sort the different raw poultry portions without physical handling by human operators is of the utmost importance (Bardic, 2004).

The use of intelligent systems based on neural networks, as well as image processing techniques to improve food production, processing, and quality in general and poultry processing in particular, has accelerated over the past decade (Khashman, 2012; Khashman and Mamedov, 2011; Khashman and Asiksoy, 2010; Balasubramaniana *et al.*, 2009; Blasco *et al.*, 2007, 2009; Kashaninejad, Dehghani and Kashiri, 2009; Marquez *et al.*, 2009; Moral *et al.*, 2009; Moreda *et al.*, 2009; Nakariyakul and Casasent, 2009; Singh *et al.*, 2009; Xie, Ying and Ying, 2009; Chegini *et al.*, 2008; Jarimopas and Jaisin, 2008; Lertworasirikul and Tipsuwan, 2008; Wu *et al.*, 2008; Chao *et al.*, 2007; Du and Sun, 2006; Park *et al.*, 2006; Yang, Chao and Chen, 2005). These works demonstrated successful applications of computer vision technologies and neural network systems to food production and processing. All these systems share a common objective, which is to improve the production yield while maintaining minimal costs.

The work presented within this chapter shares this common objective with the above previous works, as well as addressing the labour cost and potential health hazard problems by eliminating the physical contact between human labourers and raw poultry meat.

28.3 Intelligent poultry portion identification

In this chapter, we address the problem of sorting or physically separating cut raw chicken portions as they are run on a conveyor belt in an assumed poultry processing plant. We describe a prototype model design for an automated poultry portion sorting system based on using computer vision and artificial intelligence. Our novel approach simulates a human worker's task as he/she identifies the portions on the conveyor belt, bearing in mind that the human worker is able to identify a portion regardless of its size, appearance and orientation on the belt. The proposed identification system is inspired by the idea of replacing manual portion sorting by human labourers, while modelling the way the human sorter performs portion identification.

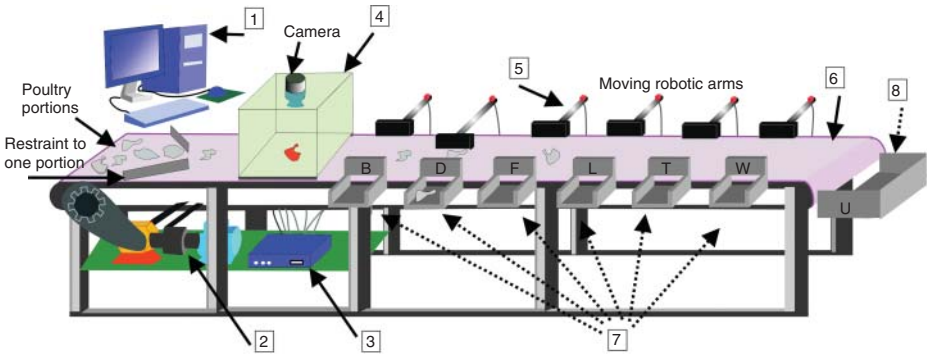


Figure 28.1 Automated poultry portion sorting system prototype (1, user interface; 2, conveyor power drive; 3, control unit; 4, poultry portion image grabbing box with digital camera and lighting source; 5, robotic arms for portion segregation; 6, conveyor belt; 7, containers for sorted portions: B (breast), D (drumstick), F (fillet), L (leg), T (thigh) and W (wing); 8, unidentified portions container). *See plate section for colour version*

For this purpose, we built an image database for implementing the proposed system using six raw chicken portions, namely breast, drumstick, fillet, leg, thigh and wing. Moreover, the proposed identification system is designed to be rotational invariant, which means that the chicken portions should be correctly identified regardless of their orientation (rotation) as they fall on to the conveyor belt. This is motivated by the fact that in a poultry processing plant, the cut-up portions may land on the conveyor belt at random and at different angles or orientations. To achieve the rotational invariance property, each of the six chicken portions was manually rotated in our lab by 15 degrees and its images were captured. The output of this identification system can be used for further processing such as moving mechanical robotic arms to physically separate the different portions into separate containers. Figure 28.1 shows a prototype of the fully automated poultry sorting system.

The development of such a system can be carried out in three stages. The first is the poultry portion image database construction. In this work, we consider chicken portions; however, the same process can be applied to identify other birds. Second, the collected database images undergo digital image processing in order to prepare the training and testing data for the artificial neural network classifier. Finally, the neural network model is trained to identify the portions using some of the database images, while maintaining the remaining images for testing and validation purposes. In this section we describe each of these stages in detail.

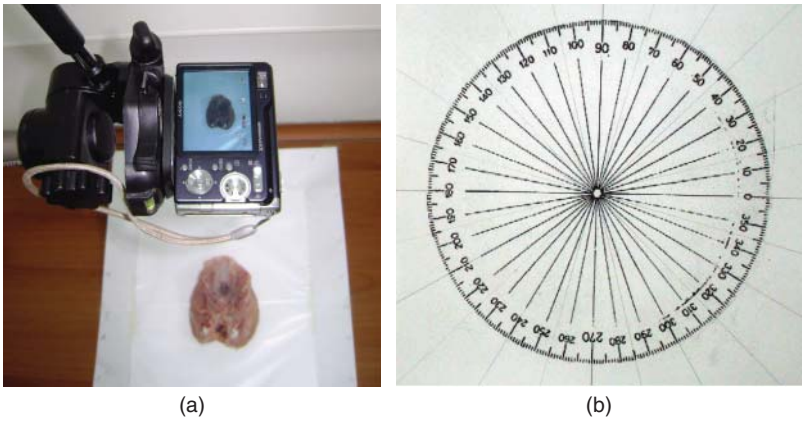
28.3.1 Image acquisition and database construction

During this stage, various images of the raw bird's portions are captured. For the purpose of this work, we used a medium-size raw chicken weighing

approximately 1.7 kg. The chicken was cut up manually into several portions and images of the most commonly preferred chicken portions by consumers were captured; these were: breast, drumstick, fillet, leg, thigh and wing. The process of acquiring the images involved laying the chicken portions against a white background, in order to model a white conveyor belt in a poultry processing plant. The distance between the camera lens and the chicken portion was kept constant at 70 cm throughout the image acquisition process, as shown in Figure 28.2a.

Images of the obverse (top) sides and reverse (bottom) sides of each of the six chicken portions were captured under fluorescent lighting as colour images, which were then converted to greyscale in order to reduce computational expenses. Capturing the portion obverse and reverse sides was in consideration of the possibility that in a processing plant a portion may land on a conveyor belt with its obverse or reverse side facing the camera. Moreover, in order to assure the rotational invariance property of the identification system, several images of each portion were also captured with the portion being rotated by intervals of 15° (see the rotation scale in Figure 28.2b), thus resulting in 48 images (24 obverse and 24 reverse) of each of the six portions; this provides us in total with 288 chicken portion images to be used in the implementation of this work. Figure 28.3 shows examples of the six different chicken portions in colour and greyscale images prior to further image pre-processing. Figure 28.4 shows the images obtained due to 15° rotations of a chicken portion's obverse sides.

The image database that is used for the development and implementation of our intelligent chicken portion identification system has now a total of 288 images, representing equally the six different chicken portions.



(a) (b)

Figure 28.2 (a) Image capturing stage of raw chicken portions and (b) portion orientation scale. *See plate section for colour version*

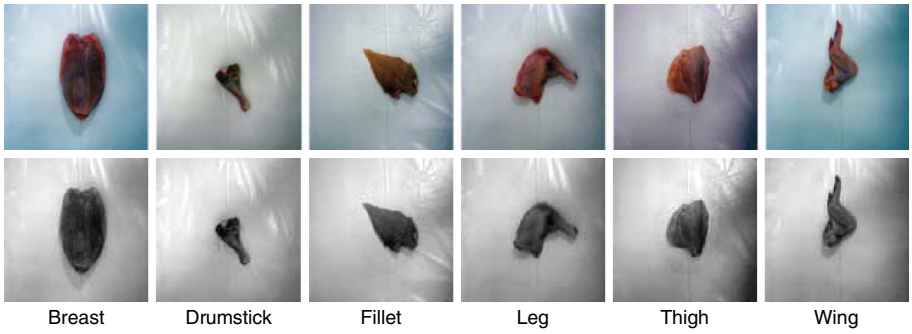


Figure 28.3 Examples of captured colour images of the six chicken portions and their greyscale versions. *See plate section for colour version*

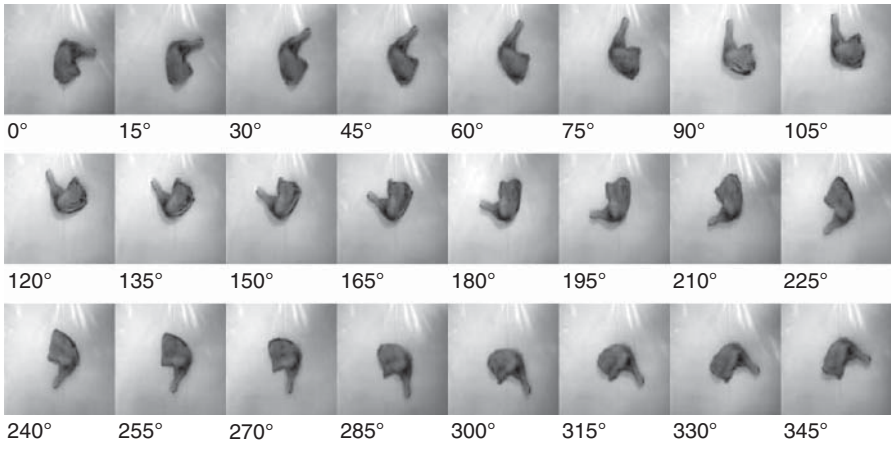


Figure 28.4 Obverse side images of a chicken leg's rotations by 15° during image database construction

28.3.2 Image processing phase

During this stage, images of the chicken portions undergo colour conversion to greyscale, resizing and feature extraction via pattern averaging in preparation to be presented to the final processing stage, which is training a neural network model to identify the different portions. Once the neural network is trained, the final stage involves only a single feed forward computation of the trained neural network final parameters to identify a newly presented chicken portion image. If the test portion of the image does not belong to the defined portion type (six types in this work), then the system's response would be 'unknown portion'. Therefore, this image processing phase can be considered as a data preparation stage for implementing the neural network identifier.

Consequently, care must be taken in order to provide the neural network with sufficient data representations of the rotated chicken portions if we are to achieve meaningful learning and yet maintain low computational costs.

The captured two-dimensional red–green–blue (RGB) colour images of the rotated chicken portions are of size 2592×1944 pixels. Using the Adobe Photoshop 7.0 Element software tool, these images are converted to greyscale and then resized to 100×100 pixels with pixel values between 0 and 255.

Feature extraction via pattern averaging is then applied to the greyscale 100×100 pixel images. Feature extraction here refers to the process of obtaining numerical vector representations of regions within the image; these vectors are used as inputs to the neural network in the third stage. A portion of image is segmented using 5×5 kernels or masks, and the pixel values within each kernel are averaged and saved as feature vectors for feeding the neural network. This method results in a 20×20 'fuzzy' bitmap, which represents the different chicken portions at various rotations. Other segment sizes can also be used; however, the larger the segment size, the higher the computational cost will be. A 5×5 segment size results in a 20×20 feature vector bitmap, thus requiring 400 neurons in the neural network input layer. Averaging the segments within an image reduces the amount of data required for neural network implementation, thus maintaining low computational costs and faster identification. Pattern averaging can be defined as follows:

$$PatAv_i = \frac{1}{s_k s_l} \sum_{l=1}^{s_l} \sum_{k=1}^{s_k} p_i(k, l) \tag{28.1}$$

where k and l are segment coordinates in the x and y directions, respectively, i is the segment number, S_k and S_l are the segment width and height, respectively, $P_i(k, l)$ is the pixel value at coordinates k and l in segment i and $PatAv_i$ is the average value of the pattern in segment i that is presented to neural network input layer neuron i. The number of segments in each image of size XY pixels ($X = Y = 100$) containing a chicken portion, as well as the number of neurons in the input layer, is i, where $i = 1, 2, 3, \ldots, n$, and

$$n = \left(\frac{X}{s_k}\right)\left(\frac{Y}{s_l}\right) \tag{28.2}$$

Using this pre-processing method in previous studies, Khashman (2008) showed sufficient representation of different objects within tested images, where meaningful data within the averaged patterns were obtained to aid the neural network learning and classification. Additionally, pattern averaging marginally reduces the processing time and, for the work presented here, pattern averaging also overcomes the problem of varying pixel values within the segments as a result of different rotations; thus it provides the rotational invariant property of the system.

28.3.3 Neural network arbitration phase

This is the final stage in operating the proposed identification system. Here a neural network classifier is trained using the averaged patterns (feature vectors) obtained from the previous image processing phase. Once the neural model is trained, this phase will only comprise generalizing the trained neural model using one forward pass, starting with an image of a poultry portion and ending with identifying the portion.

Neural model algorithm With the existence of many different neural network models, the choice of which model to employ for this system is important. The neural model should be efficient, fast and yet simple to train and re-train again if required. For these reasons, we opt to use the popular supervised learner and the back-propagation learning algorithm (Rumelhart, Hinton and Williams, 1986).

The implementation of this neural model requires a software environment to simulate the arbitration process, where input image examples are 'shown' to the neural model and its identity is 'told'. Repetition of this type of supervised training is how these neural models learn. We stop training them once their 'guessing' of the identity of many input images returns a minimal error value.

The algorithm equations that describe this neural model training and operation can be divided into two categories. First, the *feed-forward calculations* are used in both the training mode and in the operation of the trained neural network. Second, the *error back-propagation calculations* are applied only during training. A detailed description of the back-propagation algorithm equations can be found at many available online resources, for example Khashman (2013).

Neural model design Our neural network model has three layers with 400 input neurons, 30 hidden neurons and 6 output neurons. The number of neurons in the input layer is dictated by the number of averaged segments in the 20×20 bitmap for each chicken portion image. The choice of 30 neurons in the hidden layer was a result of various training experiments using lower and higher hidden neuron values. The chosen number assured meaningful training while keeping the time cost to a minimum. The six neurons in the output layer represent the six chicken portions: breast, drumstick, fillet, leg, thigh and wing. An output binary code is used to describe each identified portion, as shown in Table 28.1.

The activation function used for the neurons in the hidden and output layers is the sigmoid function. During the learning phase, initial random weights of values between -0.3 and 0.3 were used. The learning rate and the momentum rate were adjusted during various experiments in order to achieve the required minimum error value of 0.005, which was considered as sufficient for this application. Figure 28.5 shows the topology of this neural network.

Table 28.1 Identified chicken portion representation as binary code at the neural network output layer

Chicken portion	Binary code representation
Breast	1 0 0 0 0 0
Drumstick	0 1 0 0 0 0
Fillet	0 0 1 0 0 0
Leg	0 0 0 1 0 0
Thigh	0 0 0 0 1 0
Wing	0 0 0 0 0 1

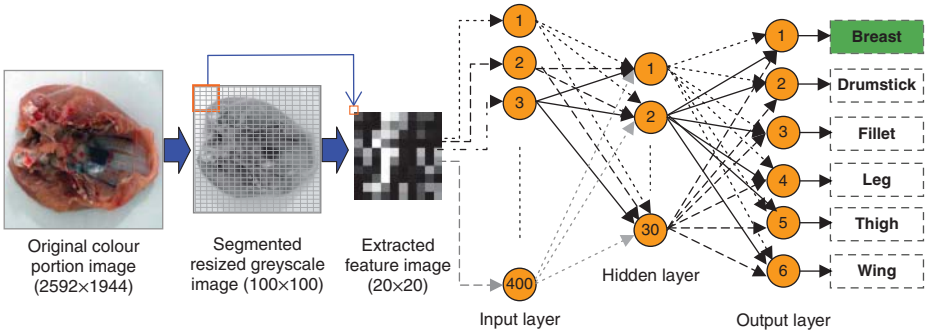


Figure 28.5 The intelligent identification system: image processing and neural network classification. *See plate section for colour version*

Neural identifier training and testing In order to train and test the neural network within the identification system, the image database is divided into two sets: the training set and the testing set. The training set images are presented to the neural network repeatedly during training, whereas the testing set images are not presented to the neural network prior to completion of training. The correct identification of the testing set images signifies the neural system's capability in a real-life implementation. In our recent studies on improving the intelligent system for raw poultry portion identification, we explored using clean and noisy portion images (Khashman and Asiksoy, 2010) and explored different neural network learning schemes (Khashman, 2012; Khashman and Mamedov, 2011). In this chapter, we describe what we consider the optimal learning scheme and setup for a neural network identifier. However, we need to clarify in the beginning what is meant by learning schemes.

The motivation of using different schemes is to investigate the capability of the neural system in identifying the different chicken portions at different orientations, while being trained with minimal training data; the aim is to keep the time costs as low as possible. The main differences between different

Table 28.2 Optimal scheme of neural network performance and final parameters by using a 2.8 GHz PC with 2 GB of RAM, Windows XP OS and Borland C^{++} compiler (CIR, correct identification rate)

Input neurons	400
Hidden neurons	30
Output neurons	6
Learning rate	0.0031
Momentum rate	0.29
Minimum error	0.005
Iterations	2645
Training time (seconds)	220.06
Run time (seconds)	4.31×10^{-4}
CIR – training images	(144/144) 100%
CIR – testing images	(143/144) 99.31%
CIR – overall	(287/288) 99.65%

learning schemes are the training-to-testing data ratios and the chosen training images. Considering that our database contains a total of 288 raw portion images, a data ratio dictates how many of these images are used for training (repeated exposure to the neural network) and how many are used for testing (shown once after training to measure the model's training success).

Here we opt for an optimal learning scheme where 50% of the database images are used for training and the remaining 50% for testing, that is (1:1) training-to-testing data ratio or learning ratio. Under this learning scheme and using learning and momentum coefficients of 0.0031 and 0.29, respectively, a correct identification rate (CIR) of 100% was obtained for the training image set (144 images). Using the testing image set (144 images not previously exposed to the neural network), the CIR was 99.31%, that is recognizing correctly 143 out of the 144 portions. Table 28.2 lists the final parameters for an optimized neural network learning to identify raw chicken portions. Figure 28.6 shows the learning curve of the neural network during training.

There was only one incorrect identification (out of 288), which was identifying a wing image as that of a drumstick (see Figure 28.7). However, we consider the neural model identifier to be very successful due to the high overall correct identification rate (99.65%) and its time cost-effective operation. Thus, the robustness, flexibility and speed of this identification system have been demonstrated through this application.

28.4 Future possible applications

The practical application of this work can be realized in any poultry processing plant where complete automation is required. The output of the described intelligent sorting system can be further used to move robotic arms to physically separate and sort the cut-up poultry portions. The system does

Figure 28.6 The learning curve of the neural network identifier during training

Figure 28.7 The only incorrect identification of a chicken portion

not depend on weight of portions, but rather mimics the way a human worker would identify a chicken portion using a camera as an input device and an artificial neural network as the decision maker.

We trained the system in this work to recognize the main chicken portions, but other portions and other poultry birds can also be identified, including their portion images in the training phase. This work differs from existing systems as it relies totally on the shape and coarse texture of a portion, using images as input data, and discards information like weight or size of the portion. Another advantage is the elimination of the need for using many birds during the development of the system; in fact, training the system can be achieved using only one bird and its portions.

28.5 Conclusions

This chapter presented a prototype model design for an intelligent poultry portion sorting system. The prototype model can be used in a poultry

processing plant to automatically sort or separate cut-up poultry portions into designated separate containers prior to packaging. Our concept is to fully automate this sorting process, which is usually carried out by human workers in poultry processing plants. Apart from cutting down the labour costs and potential human errors, such a sorting system completely eliminates physical contact and thus potential contamination between raw poultry parts and human workers.

In order to achieve these objectives, we employ in our approach an artificially intelligent neural network model and basic image processing techniques with the aim of mimicking the way a human worker would identify poultry portions passing on a conveyor belt. The output of our proposed approach to automate the sorting process is a decision on what the portion is, that is portion identification. This decision, which is in binary code, can be potentially used to drive different robotic arms across the conveyor belt in order to 'push' or move an identified portion into its designated container. In this work, we used raw chicken as the bird and focused on training the system on the main popular portions often purchased by us. These were: chicken breast, drumstick, fillet, leg, thigh and wing. In our design, the portions that are not identified and sorted end up in a separate container labelled 'unidentified portion'. These could be re-processed again by the automated system or can be allocated for a different use.

The implementation of our proposed sorting system achieved an astonishing overall correct identification rate of 99.65% of the chicken portions. The identification time for one portion was also fast to within a fraction of a second. Therefore, we consider such an approach ideal to manual poultry portion sorting. In fact, based on our proposed system and providing training, it is possible that any manual product sorting system can be automated following this approach. To summarize our approach to this problem, we may divide the system into three stages: product image database construction, image processing and neural network training/testing. For this work, we look forward soon to add more portions, such as the whole bird and other portions such as neck, tail, saddle, skeleton frame of the breast and upper back.

References

Balasubramaniana, S., Panigrahi S., Logue, C.M., Gu, H. and Marchello, M. (2009) Neural networks-integrated metal oxide-based artificial olfactory system for meat spoilage identification. *Journal of Food Engineering*, **91**, 91–98.

Bardic, A. (2004) Poultry in motion: demand for labor-saving automated poultry processing equipment escalates. *National Provisioner*, pp. 1–8.

Blasco, J., Aleixos, N., Gómez, J. and Molto, E. (2007) Citrus sorting by identification of the most common defects using multispectral computer vision. *Journal of Food Engineering*, **83**, 384–393.

Blasco, J., Cubero, S., Sanchís, J.G., Mira P. and Moltó, E. (2009) Development of a machine for the automatic sorting of pomegranate (*Punica granatum*) arils based on computer vision. *Journal of Food Engineering*, **90**, 27–34.

Chao K., Chen, Y.R., Ding, F., Yang, C.-C. and Chan, D.E. (2007) Development of two-band color-mixing technique for identification of broiler carcass conditions. *Journal of Food Engineering*, **80**, 276–283.

Chegini, G.R., Khazaei, J., Ghobadian, B. and Goudarzi, A.M. (2008) Prediction of process and product parameters in an orange juice spray dryer using artificial neural networks. *Journal of Food Engineering*, **84**, 534–543.

Du, C.J. and Sun, D.W. (2006) Learning techniques used in computer vision for food quality evaluation: a review. *Journal of Food Engineering*, **72**, 39–55.

Gainco Inc. (2011a) http://www.gainco.com. Accessed August 2011.

Gainco Inc. (2011b) http://www.gainco.com/classifier. Accessed August 2011.

Jarimopas, B. and Jaisin, N. (2008) An experimental machine vision system for sorting sweet tamarind. *Journal of Food Engineering*, **89**, 291–297.

Kashaninejad, M., Dehghani, A.A. and Kashiri, M. (2009) Modeling of wheat soaking using two artificial neural networks (MLP and RBF). *Journal of Food Engineering*, **91**, 602–607.

Keener, K.M., Bashor, M.P., Curtis, P.A., Sheldon, B.W. and Kathariou, S. (2004) Comprehensive review of *Campylobacter* and poultry processing. *Comprehensive Reviews in Food Science and Food Safety*, **3**, 105–116.

Khashman, A. (2008) Blood cell identification using a simple neural network. *International Journal of Neural Systems*, **18**, 453–458.

Khashman, A. (2012) Automatic identification system for raw poultry portions. *Journal of Food Process Engineering*, **35**, 727–734.

Khashman, A. (2013) Online at: http://staff.neu.edu.tr/~amk/Khashman_NN_BP_Handout.pdf.

Khashman, A. and Asiksoy, G. (2010) Identification of noisy poultry portion images using a neural network. *Proceedings of 9th WSEAS International Conference on Artificial Intelligence, Knowledge Engineering, and Databases (AIKED'09)*, University of Cambridge, England.

Khashman, A. and Mamedov, F.S. (2011) Automation of poultry sorting plant using neural networks. *Proceedings of the International Symposium on Advances in Applied Mechanics and Modern Information Technology*, Baku, Azerbaijan.

Lertworasirikul, S. and Tipsuwan, Y. (2008) Moisture content and water activity prediction of semi-finished cassava crackers from drying process with artificial neural network. *Journal of Food Engineering*, **84**, 65–74.

Marquez, A.J., Herrera, M.P.A., Ojeda, M.U. and Maza, G.B. (2009) Neural network as tool for virgin olive oil elaboration process optimization. *Journal of Food Engineering*, **95**, 135–141.

Moral, F.G., Guillén, A., Moral, L.G., O'Valle, F., Martínez, L., Moral, R.G. (2009) Duroc and Iberian pork neural network classification by visible and near infrared reflectance spectroscopy. *Journal of Food Engineering*, **90**, 540–547.

Moreda, G.P., Ortiz-Cañavate, J., García-Ramos, F.J. and Ruiz-Altisent, M. (2009) Non-destructive technologies for fruit and vegetable size determination. *Journal of Food Engineering*, **92**, 119–136.

Nakariyakul, S. and Casasent, D.P. (2009) Fast feature selection algorithm for poultry skin tumor detection in hyperspectral data. *Journal of Food Engineering*, **94**, 358–365.

Park, B., Lawrence, K.C., Windham, W.R. and Smith, D.P. (2006) Performance of hyperspectral imaging system for poultry surface fecal contaminant detection. *Journal of Food Engineering*, **75**, 340–348.

Rumelhart, D., Hinton, G.E. and Williams, R.J. (1986) Learning internal representations by error propagation. In: *Parallel Distributed Processing*, D.E. Rumelhart and R.J. McClelland (eds). MIT Press, pp. 318–362.

Sams, A.R. (2001) *Poultry–Meat Processing*. CRC Press, Boca Raton, Florida, USA, pp. 19–34.

Singh, R., Ruhil, A.P., Jain, D.K., Patel, A.A. and Patil, G.R. (2009) Prediction of sensory quality of UHT milk – a comparison of kinetic and neural network approaches. *Journal of Food Engineering*, **92**, 146–151.

Somsen, D., Capelle, A. and Tramper, J. (2004) Production yield analysis in the poultry processing industry. *Journal of Food Engineering*, **65**, 479–487.

Van Hoogen, G.J. (2005) Poultry processing: developing new tools to be competitive. *Proceedings of XVIIth European Symposium on the Quality of Poultry Meat*, pp. 282–284.

Wu, D., Yang H., Chen, X., He, Y. and Li, X. (2008) Application of image texture for the sorting of tea categories using multi-spectral imaging technique and support vector machine. *Journal of Food Engineering*, **88**, 474–483.

Xie, L., Ying, Y. and Ying, T. (2009) Classification of tomatoes with different genotypes by visible and short-wave near-infrared spectroscopy with least-squares support vector machines and other chemometrics. *Journal of Food Engineering*, **94**, 34–39.

Yang, C.C., Chao, K. and Chen, Y.R. (2005) Development of multispectral image processing algorithms for identification of wholesome, septicemic, and inflammatory process chickens. *Journal of Food Engineering*, **69**, 225–234.

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Figure 4.5 Various extruded products like expanded corn balls (top), extruded-toasted corn chips (middle) and texturized vegetable protein (TVP) (bottom)

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Figure 5.4 Conventional (left) and air-incorporated (right) gellan gels

Figure 6.3 Modern-day horizontal batch retort (source: Photo courtesy of JBT FoodTech, formerly FMC FoodTech, Madera, CA)

Figure 6.4 Batch retort system showing battery of retorts in a large cook room operation (source: Photo courtesy of JBT FoodTech, formerly FMC FoodTech, Madera, CA)

Figure 6.6 Exterior view of a continuous hydrostatic sterilizer (source: Photo courtesy of JBT FoodTech, formerly FMC FoodTech, Madera, CA)

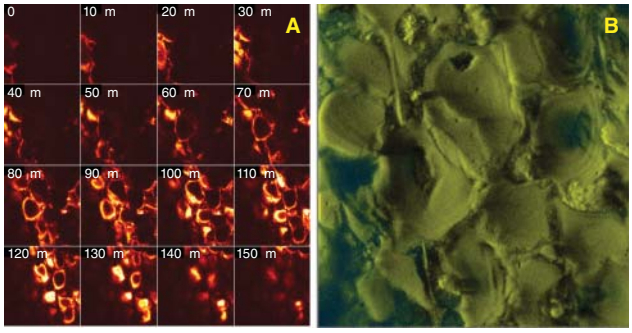


Figure 9.3 (A) Confocal images in the fluorescence mode of oil distribution in a potato chip fried in stained oil (170 °C, 3 min) observed at ×20. (B) Three-dimensional reconstruction from the serial sections in Figure 9.2 using Imaris software (source: Pedreschi *et al*. 1999. Reproduced with permission of John Wiley & Sons, Ltd)

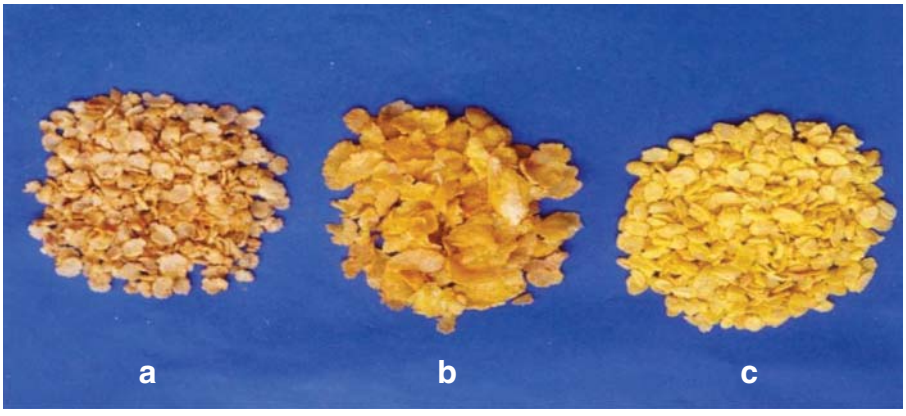


Figure 10.4 Breakfast cereals from (a) sorghum, (b) corn and (c) rice

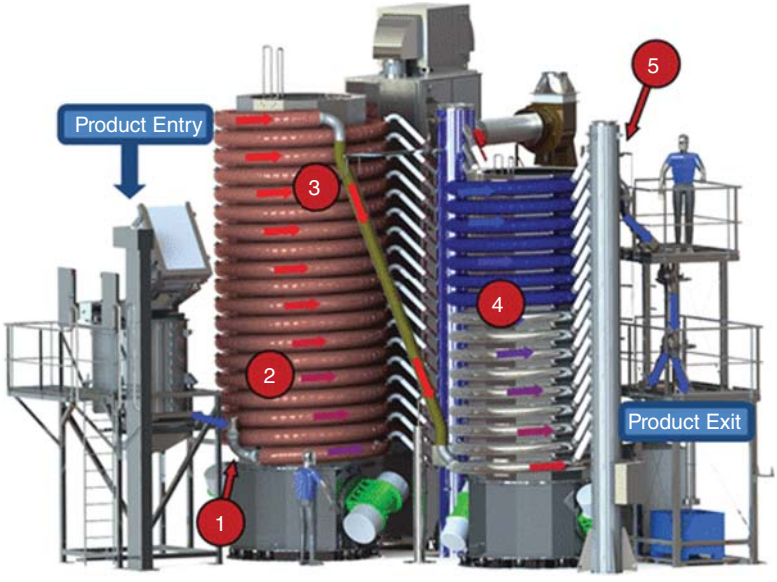


Figure 10.8 Continuous spiral roaster: 1, feeding system, 2, vibratory hot tubes to transport materials, 3, roasted materials progressing tubes, 4, cooling tubes and 5, product exit (source: REVTECH, France. Reproduced with permission of REVTECH Process Systems)

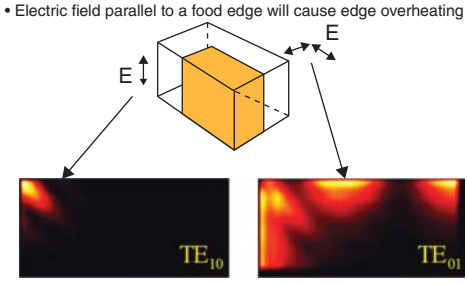


Figure 18.2 Illustration of edge overheating, where the electric field parallel to a food edge will cause edge overheating (source: Sundberg, 1998, Reproduced with permission of SIK, Sweden)

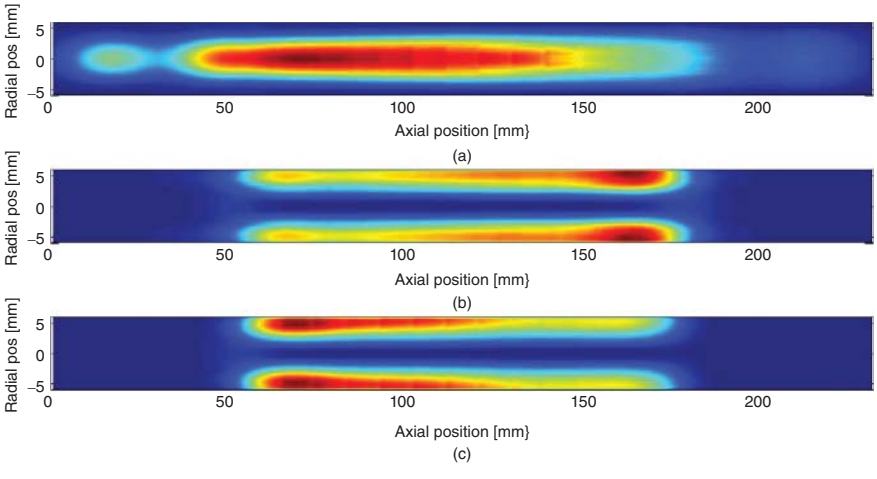
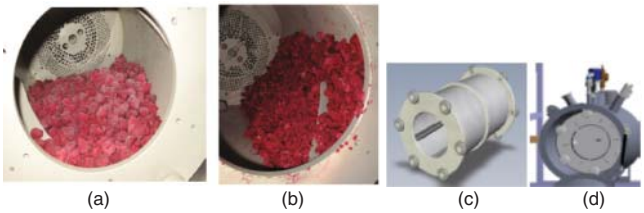


Figure 18.4 Dissipated power in the load of a tubular microwave sterilisation process (pos stands for position) (source: Isaksson, 2013, Reproduced with permission of SIK, Sweden)



Brix	8–13%
Initial weight	5.kg (83% moisture content)
End weight	0.82 kg (7–10% moisture content)
Drying time	3.4 hours
Max temperature	35°C
Applied micro wave energy	4.64 kW

Figure 18.5 Microwave freeze dryer (a), (b), based on a closed rotating drum (c), (d), for preservation of raspberries (source: Püschner 2013. Reproduced with permission of Püschner)

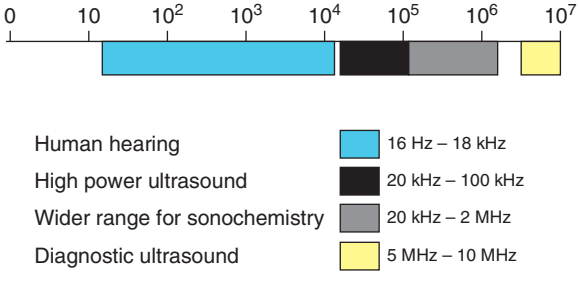


Figure 21.1 Range of ultrasound frequency (source: Leonelli and Mason, 2010. Reproduced with permission of Elsevier)

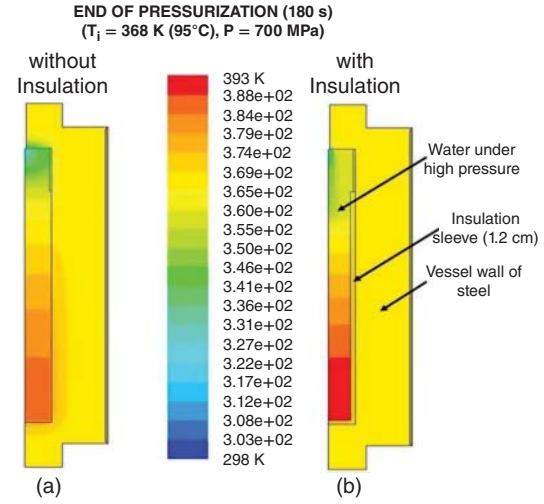


Figure 24.4 Isotherms in water and stainless steel vessel for vertical vessel at $T_i = 368\,\text{K}$, $P = 700\,\text{MPa}$, $T_{\text{inlet}} = 298\,\text{K}$, $Q = 860\,000\,\text{W/m}^3$ for $(0 \leq t \leq 180\,\text{s})$, (a) without insulation and (b) with insulation (12.7 mm thick) at the end of pressurization (180 s) (source: Khurana 2012. Reproduced with permission)

Figure 26.5 PEF units developed by DIL and the related maximum capacities; left side: pulse transformer setup; right side: semiconductor-based Marx generator equipment (source: DIL e.V., Quakenbrueck, Germany)

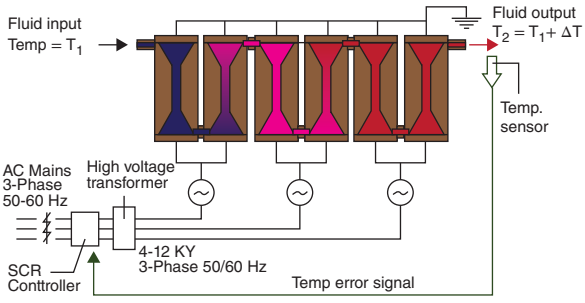


Figure 27.4 Commercial Ohmic heating processing system (source: presentation by David Reznik for the Aseptipak Global Forum, 2001, Chicago, USA. Image reproduced with kind permission from Raztek, California, USA)

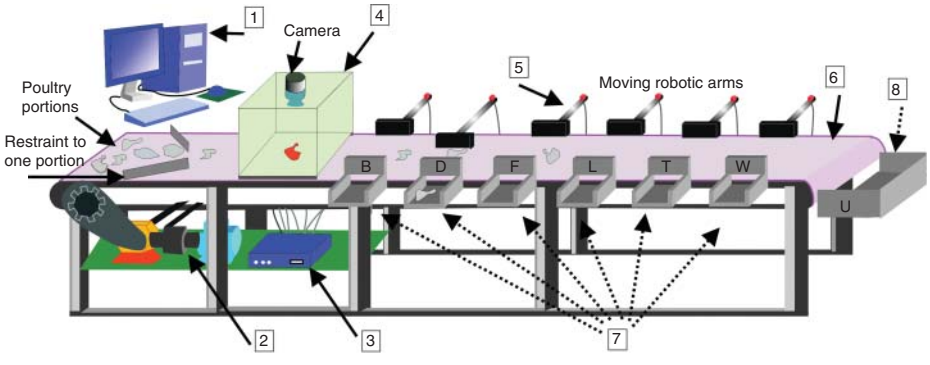
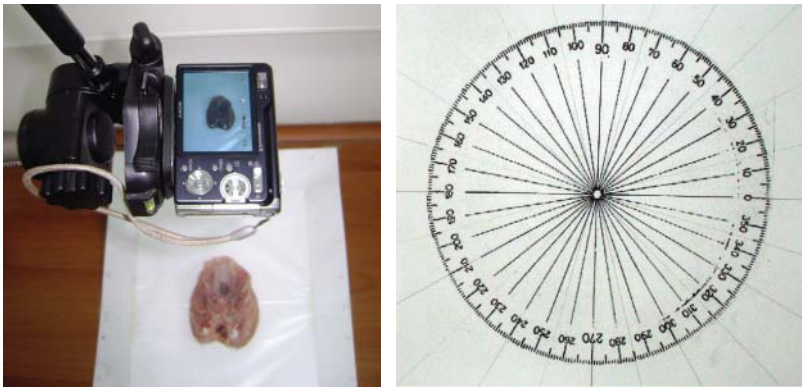


Figure 28.1 Automated poultry portion sorting system prototype (1, user interface; 2, conveyor power drive; 3, control unit; 4, poultry portion image grabbing box with digital camera and lighting source; 5, robotic arms for portion segregation; 6, conveyor belt; 7, containers for sorted portions: B (breast), D (drumstick), F (fillet), L (leg), T (thigh) and W (wing); 8, unidentified portions container)



(a) (b)

Figure 28.2 (a) Image capturing stage of raw chicken portions and (b) portion orientation scale

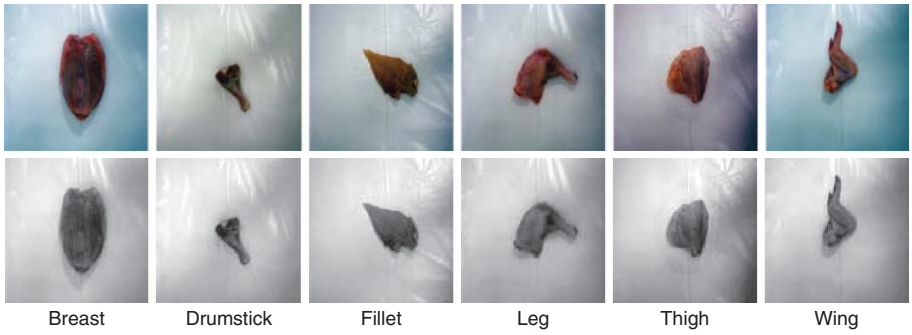


Figure 28.3 Examples of captured colour images of the six chicken portions and their greyscale versions

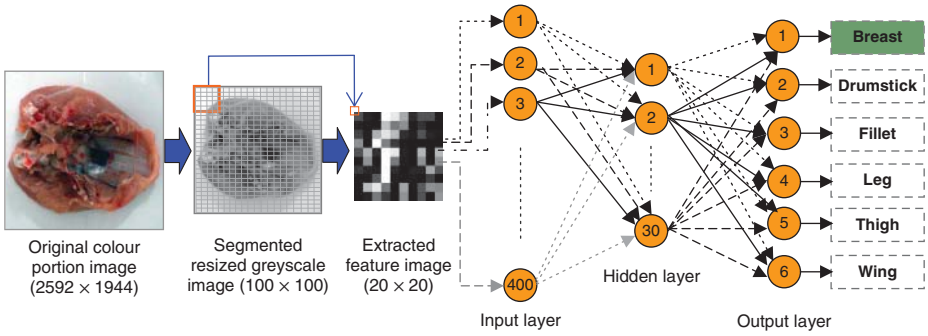


Figure 28.5 The intelligent identification system: image processing and neural network classification

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