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Preface

The management of wastes, in particular of industrial waste, in an economically and environmentally acceptable manner is one of the most critical issues facing modern industry, mainly due to the increased difficulties in properly locating disposal works and complying with even more stringent environmental quality requirements imposed by legislation. The development of innovative systems to maximize recovery of useful materials and/or energy in a sustainable way has become necessary. The significant residual waste streams from pulp and paper mills include wastewater treatment sludges, lime mud, lime slaker grits, green liquor dregs, boiler and furnace ash, scrubber sludges and wood processing residuals. Pulp and paper mill industries are always associated with disposal problem of highly contaminated sludge or bio-solids. In countries with large scale Pulp and paper production, the huge amount of waste generated has prompted the government and industries to find new use of these bio-solids. Paper mill sludges have a net environmental advantage over sewage sludges in that they are nearly pathogen free; handling and use pose lower health risks. Land filling, land application, composting, land-spreading to improve soil fertility, production of ethanol and animal feed, pelletization of sludge, manufacture of building and ceramic materials and lightweight aggregate, landfill cover barrier are among the waste management options studied. The challenge to find efficient methods for firing sludge still exists today and is becoming increasingly important as pulp and paper mill strive to be competitive. So far, incineration has been the primary alternative to landfill. However, incineration is associated with environmental pollution problems. The emission of gaseous NO_x and SO_2 are the major precursors of acid rain. The residue ash contains various toxic metals which need to be landfilled and hence result in ground water contamination. The plastics and glue found in the sludge are the sources of chlorinated compounds such as HCl, dioxins and furans which are major threat to the environment. This book presents general introduction on waste management in pulp and paper industry, generation of waste in pulp and paper mills, waste composition, methods of sludge pretreatment, processes and technologies for conversion of pulp and paper mill waste into valuable products, state-of the-art waste reduction techniques employed in the pulp and paper industry worldwide and future trends.

Patiala, India Pratima Bajpai
June 2014

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Abbreviations

AOX	Adsorbable organic halides
BOD	Biochemical oxygen demand
CCPs	Coal combustion products
CDs	Cyclodextrins
CEC	Cation exchange capacity
COD	Chemical oxygen demand
CTMP	Chemithermomechanical pulp
DIP	Deinked pulp
DS	Dry solids
DSC	Dry solids content
EPA	Environment Protection Agency
ESP	Electrostatic precipitator
FRCA	Fine recycled concrete aggregate
GHG	Greenhouse gas
HPSEC	High pressure size exclusion chromatography
IPPC	Integrated pollution prevention and control
LHL	Low-high-low temperature
MDF	Medium density fiberboard
MDI	Methylene diphenyl diisocyanate
MTCI	Manufacturing and Technology Conversion International
NSSC	Neutral sulfite semi-chemical
OFS	Oil-from-sludge
PPMB	Pulp and paper mill biosolids
RPS	Recycled paper sludge
RTP	Rapid thermal processing
SCWG	Supercritical water gasification
SCWO	Supercritical water oxidation
SHF	Separate hydrolysis and fermentation
SSCF	Simultaneous saccharification and co-fermentation

SSF	Simultaneous saccharification and fermentation
UFA	Unsaturated fatty acids
WAO	Wet air oxidation
WPSA	Waste paper sludge ash
WWTP	Wastewater treatment plant
WWTS	Wastewater treatment sludge

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Chapter 1
General Introduction

Abstract The production of paper consumes high quantities of energy, chemicals and wood pulp. Consequently, the paper production industry produces high environmental emission levels mainly as carbon dioxide due to energy consumption, or solid waste streams which include wastewater treatment sludges, lime mud, lime slaker grits, green liquor dregs, boiler and furnace ash, scrubber sludges and wood processing residuals. In terms of volume, most solids or liquids are those from the treatment of effluents, although waste from wood is also produced in large quantities. Wastewater treatment plant residuals are the largest volume residual waste stream generated by the pulp and paper industry. The general background of waste management in pulp and paper industry are presented.

Keywords Waste management • Pulp and paper industry • Solid waste • Wastewater treatment sludges • Lime mud • Lime slaker grits • Green liquor dregs • Boiler ash • Furnace ash

1.1 The Paper and Paperboard Industry in the Global Market

The pulp and paper industry plays an integral role in the global economy. Approximately 400 million tonnes of paper and paperboard are produced globally (Bajpai 2013). Paper and paperboard consumption continues to grow in Asia, especially in China. Asia already accounts for well over a third of global paper and paperboard production. Meanwhile production in North America is shrinking. For the past 4 years, China has maintained the top spot for both demand and production of total paper and board, with the United States remaining in second place. China accounted for 25 % of world demand and 26 % of global production of total paper and board in 2012. In terms of pulp production, the United States remained the top producing country in the world with 50.4 million tonnes in 2012. China came in second producing 18.2 million tonnes. Estimates suggest that global paper consumption in 2025 will amount to 500 million tonnes, which means growth of about 1.6 % a year. Asia's share of global consumption is already 44 %. Europe and North America account for almost a third of consumption. Demand in Eastern Europe is also growing faster than in traditional markets. According to Confederation

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of European Paper Industries (CEPI) (2011), production of pulp in Europe was slightly increasing over the period of 1991–2011, while the production of paper and cardboard in European countries increased by 50 % over the same period. The production of pulp, paper and cardboard has grown in the Russian Federation as well (Deviatkin 2013). The production processes result in waste generation. According to Monte et al. (2009) in 2005 in Europe, 11 million tonnes of solid waste, including 7.7 million tonnes of waste from recycled fibre processing was generated during the production of 99.3 million tonnes of paper and in Russia in 2011, 15.2 million tonnes of pulp, paper, and board were produced (Deviatkin 2013). Meanwhile, about 25 % of all solid wastes generated in Pulp and Paper Industry and directed to landfills are wastewater treatment sludge and deinking sludge (Vuoristo 2012).

1.2 General Aspects of Waste Management

Solid waste is largely produced from pulping and deinking processes and wastewater treatment. Composition and amount of sludge are strongly affected by the raw materials used by the process, paper grade being produced, the production and wastewater treatment technologies and also the properties to be obtained (Monte et al. 2009; Abubakr et al. 1995). The important residual waste streams from pulp and paper mills include:

- Wastewater Treatment Sludges
- Lime Mud
- Lime Slaker Grits
- Green Liquor Dregs
- Boiler And Furnace Ash
- Scrubber Sludges
- Wood Processing Residuals

In terms of volume, most solids or liquids are those from the treatment of effluents, although waste from wood is also produced in huge quantities (Gavrilescu 2004, 2005; IPPC 2001; CANMET 2005; Battaglia et al. 2003; Geng et al. 2006; Suriyanarayanan et al. 2010; Krigstin and Sain 2005; Mladenov and Pelovski 2010; Springer 1993; Krogmann et al. 1997; Krogmann 1998; Charlie 1977; Hudson and Lowe 1996; Kay 2002, 2003; Reid 1997, 1998; Glenn 1997; Watson and Hoitink 1985; Nurmesniemi et al. 2007; Vehlow et al. 2007; Pickell and Wunderlich 1995). In countries with large scale Pulp and Paper production, the large amount of waste generated has prompted the government and industries to find new uses of these biosolids (Kay 2003; Glenn 1997; Watson and Hoitink 1985; Scott and Smith 1995; Abubakr et al. 1995; Vehlow et al. 2007; Young 1982; Turnbull 1982; Axegard and Backlund 2002; Ishimoto et al. 2000). In comparison to sewage sludges, paper mill sludges have a net environmental advantage in that they are almost free of pathogens; handling and use pose lower health risks.

Wastewater treatment plant residuals (WWTP) are the largest volume residual waste stream generated by the pulp and paper industry. In United States, about 5.5 million dry tons annually is produced (Bird and Talbert 2008; Thacker 2007). Wastewater treatment plant residuals are of four types:

- Primary including deinking residuals represents 40 % of WWTP residuals
- Secondary (waste activated sludge) sludge is 1 %
- Combined primary and secondary sludge is 54 %
- Dredged (5 %)

Mechanical dewatering is the norm of processing wastewater treatment plant residuals, with a solid content in the range of 30–40 % on average (Bird and Talbert 2008). When processed in this manner, the waste does not fall into the hazardous category as defined by the Resource Conservation and Recovery Act. This solid waste is low in metals, low in trace organics with low to medium nutrients. A small number of mills dry their residuals, which produce a 70–95 % solid waste rate (Thacker 2007).

Primary wastewater treatment plant residuals mostly consist of processed wood fiber and inorganic or mineral matter mainly kaolin clay, calcium carbonate, titanium dioxide. The ash (inorganic material) produced from this process ranges from less than 10 % up to 70 % (dry weight). Secondary wastewater treatment plant residuals consist mostly of non-pathogenic bacterial biomass.

As chlorinated organic compounds have a tendency to partition from effluent to solids, wastewater treatment sludge is a significant environmental concern for the pulp and paper industry. But recent trends away from elemental chlorine bleaching have reduced these hazards (Bajpai et al. 1999). A continuing apprehension is the very high pH of more than 12.5 of most residual wastes. When these wastes are disposed of in an aqueous form, they may meet the Resource Conservation and Recovery Act's definition of a corrosive hazardous waste (Bird and Talbert 2008).

The generation of sludge vary widely among mills (Lynde-Maas et al. 1997; Reid 1998; Elliott and Mahmood 2005, 2006; Krigstin and Sain 2005, 2006). EPA investigated 104 bleached Kraft mills; the sludge generation ranged from 14 to 140 kg of sludge per ton of pulp. For these 104 mills, total sludge generation was 2.5 million dry metric tons per year, or an average of approximately 26,000 dry metric tons per year per plant. Pulp making operations are responsible for generating the bulk of sludge wastes, although treatment of papermaking effluents also produces significant sludge volumes. The majority of pulp and integrated mills operate their own wastewater treatment systems and generate sludges onsite. A much larger proportion of papermaking establishments and a small number of pulp mills discharge effluents to publicly-owned wastewater treatment works.

Significant number of mills dispose sludge through land application though landfill and surface impoundment disposal are most often used for wastewater treatment sludge (Rashid et al. 2006; Bajpai et al. 1999). U.S. Department of Energy and Environmental Protection Agency consider proper land application of sludge as a beneficial use. Paper mill sludges can consume large percentages of local landfill space each year. When disposed of by being spread on cropland, concerns are raised about trace contaminants building up in soil or running off into area lakes and streams. Some pulp and paper mills burn their sludge in incinerators for onsite energy generation (CWAC; Bird and Talbert 2008).

According to a 2002 study by the American Forestry and Paper Association, wastewater treatment plant residuals were managed in United States as shown in Table 1.1 (Bird and Talbert 2008). The Confederation of European Paper Industries (CEPI) reported in 2003 that waste water treatment residuals in member countries on average were managed as shown in Table 1.2. Boiler ash and causticizing residuals were managed in United States as presented in Tables 1.3 and 1.4 respectively (NCASI 2001, 2007; Bird and Talbert 2008).

Table 1.1 Wastewater treatment plant residuals management in USA

	Wastewater treatment plant residuals (%)
Land application	14.6
Lagoon or landfill	51.8
Incineration for energy production	21.9
Other beneficial use	11.7

Based on Bird and Talbert (2008)

Table 1.2 Waste water treatment residuals management in CEPI countries

	Waste water treatment residuals (%)
Land application	37
Energy recovery	33
Landfilling	11
Other industries	19

Based on Bird and Talbert (2008), CEPI (2003)

Table 1.3 Boiler ash management in USA

	Boiler ash (%)
Land application	9.3
Landfill/lagoon	65.4
Other beneficial use	25.3

Based on Bird and Talbert (2008), NCASI (2007)

Table 1.4 Causticizing residuals management in USA

	Lime mud (%)	Green liquor dregs (%)	Slaker grits (%)
Land application	9	3	5.5
Lagoon or landfill	70	95	91
Reuse in mill	1	0	3
Other beneficial use	21	2	1

Based on Bird and Talbert (2008), NCASI (2001)

Landfilling and incineration suffer from their inherent drawback of poor economics. The reasons are presented below (Canales et al. 1994):

- The high cost associated with dewatering the sludge to 20–40 % solids or higher so as to meet the requirements of landfilling or incineration,
- The significant energy loss in evaporating the sludge-containing water in incineration or combustion of the sludges in a recovery boiler. The sludge disposal/management costs can be as high as 60 % of the total wastewater treatment plant operating costs.

Due to rapidly reducing landfill space and the secondary pollution issues associated with the conventional sludge disposal approaches and also the increasingly stringent environmental regulations, the disposal of sludges continues to be one of the major challenges for the municipal wastewater plants and most pulp and paper mills (Mahmood and Elliott 2006). This together with record high oil prices have contributed to a need to investigate methods of converting sludge waste into energy (XU and Lancaster 2008, 2009). For example, the percentage of pulp/paper sludges disposed by landfills has constantly decreased in Europe in recent years, dropping 40 % in 1990 to 20 % in 2002. In the meantime, the percentage of pulp/paper sludge used as a raw material in other industries and other applications – agriculture as soil improvers, in road construction, land reconstruction and for energy recovery has gradually increased.

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Chapter 2
Generation of Waste in Pulp and Paper Mills

Abstract In the Pulp and Paper Industry several types of solid wastes and sludge are generated. Solid waste is mainly produced from pulping, deinking processes and wastewater treatment. The waste generation is strongly affected by the production process and wastewater treatment technologies. About 40–50 kg of sludge (dry) is generated in the production of 1 tonne of paper at a paper mill and of that approximately 70 % is primary sludge and 30 % secondary sludge. The amount of sludge on a dry mass basis may vary from 20 % in a newsprint mill to 40 % in a tissue mill. The data on waste generated in pulp and paper mills and deinking mills are presented in this chapter. Waste generated through production of different paper grades from recycled fibre are also presented.

Keywords Waste generation • Pulp and paper mill industry • Solid waste • Recycled fibre • Wastewater treatment • Primary sludge • Secondary sludge

Different types of solid wastes and sludge are generated in the Pulp and Paper Industry at different production processes (Monte et al. 2009; Gavrilescu 2004, 2005; Abubakr et al. 1995). Treatment of wastewater generated at pulping, papermaking, and deinking processes is the main source of wastewater treatment sludge and deinking sludge. Tables 2.1 and 2.2 show solid waste generated in pulp and paper mills (Monte et al. 2009; IPPC 2001; CANMET 2005). Overall view of the solid waste rates in the Kraft pulp mill are presented in Table 2.3 (Gavrilescu 2004).

Balwaik and Raut (2011) have reported that about 300 kg of sludge is produced for each 1 ton of recycled paper. The amount of waste generated in paper production varies greatly within different regions, because of different recycling rates. In Finland, the ratio of recycled fiber production to paper production can be expected to be smaller than example in central Europe (Kujala 2012). This is due to the reason that most of the paper produced in Finland is exported to other countries and so the amount of recovered paper is relatively low. According to WRAP (2010), over 5 million tons of paper and board was produced in 2007. At the same time, the production of paper mill sludge from Recycled fiber production was approximately 1 million tons (Rothwell and Éclair-Heath 2007).

The generation of wastewater treatment sludge vary widely among mills (Lynde-Maas et al. 1997; Reid 1998; Elliott and Mahmood 2005, 2006;

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Table 2.1 Solid waste generated in pulp mills

Rejects
The rejects from virgin pulps consist of sand, bark and wood residues from wood handling, which are undesirable for papermaking. Rejects typically have a relatively low moisture content, significant heating values, are easily dewatered and are, generally, burned in the mill's bark boiler for energy recovery
Green liquor sludge, dregs and lime mud
These are inorganic sludges separated from the chemical recovery cycle. These sludges are normally landfilled, after dewatering and drying
Wastewater treatment sludge
It comes from two sources: primary sludge and biological sludge generated in the second clarifier. These sludges are generally blended together, a polymer added and dewatered together to a 25–40 % dry solid content
Chemical flocculation sludge
It arises from water treatment and is often transported to the landfill site due to the high content of inorganic matter and water

Based on Monte et al. (2009), IPPC (2001), CANMET (2005)

Table 2.2 Solid waste generated in paper mills

Rejects
The rejects from recovered paper are impurities and consist of lumps of fibres, staples and metals from ring binders, sand, glass and plastics and paper constituents as fillers, sizing agents and other chemicals. Rejects also have a relatively low moisture content, significant heating values, are easily dewatered and are, generally, incinerated or disposed of in landfills. Screen rejects are produced during filtration steps with screens with very small slots to remove pulp possibly containing stickies that might disturb the production process and quality of end product. Screen rejects have a high content of cellulose fibre
Deinking sludge
This residue contains mainly short fibres or fines, coatings, fillers, ink particles (a potential source of heavy metals), extractive substances and deinking additives. It is normally reused in other industries (e.g. cement, ceramics), or is incinerated, even though it has a poor heating value. Deinking sludge is generated during recycling of paper (except for packaging production). Separation between ink and fibres is driven by "flotation" process, where foam is collected on the surface of flotation cells. The generated deinking sludge contains minerals, ink and cellulose fibres (that are too small to be withhold by filters)
Primary sludge
This sludge is generated in the clarification of process water by kidney treatments, e.g. dissolved air flotation. The sludge consists of mostly fines and fillers depending on the recovered paper being processed and it is relatively easy to dewater. This sludge can be reincorporated into the process for board industry, but for high grade products can be incinerated, dumped or, otherwise, mixed with deinking or secondary sludge
Secondary or biological sludge
This sludge is generated in the clarifier of the biological units of the wastewater treatment, and it is either recycled to the product (board industry) or thickened, dewatered and then incinerated or disposed of in landfill. Secondary sludge volumes are lower than those corresponding to the primary sludge, since most of the heavy, fibrous or inorganic solids are removed in the primary clarifier. Secondary sludges are often difficult to handle (due to a high microbial protein content), and such solids need to be mixed with primary sludge to permit adequate dewatering

Based on Monte et al. (2009), IPPC (2001), CANMET (2005)

Table 2.3 Generation of waste in a Kraft mill

Waste	Yield (kg/t o.d. pulp)
Wood wastes:	
Sawdust coming from the slasher deck	10–30
Bark falling from the debarking drum	100–300
Pins and fines from chip screening	50–100
Wood waste from woodyard	0–20
Knots from pulp deknotting	25–70
Sodium salts from recovery boiler	5–15
Dregs and grit from causticizing:	5–10
Dregs	10–30
Grit	15–40
Total:	220–615

Based on Gavrilescu (2004)

Table 2.4 Generation of waste from few European pulp and paper mills

	SCA	Norske Skog	Stora Enso	Holmen
Mill production (millions of tonnes)	9,9	4,8	15,1	2,3
Total waste generated (kg/ton product)	163	163	155 (dry)	160
Recovered waste (kg/ton product)	115	138	–	136
Waste sent to landfill (kg/ton product)	47	16	22	23 (wet)
Hazardous waste (kg/ton product)	0,3	1,5	0,3	0,2

Based on Monte et al. (2009)

Table 2.5 Generation of waste in different processes in Europe

Process	Kraft	Sulphite	Mechanical semi-chemical	Recycled fibre
Specific waste (kg/Adt)	100	80	60	185
Waste generated through paper production (million tonnes)	2,1	0,2	0,8	7,7

Based on IPPC (2001), Monte et al. (2009)

Krigstin and Sain 2005, 2006; Monte et al. 2009; Abubakr et al. 1995). Not much data is available on total waste generation. This is due to the fact that most of the pulp and paper mills already have processes applied to internally treat the wastes which reduce the generation of solid waste. This applies to bark residues from debarking which are incinerated in the bark boiler and, as a result, only ashes remain as waste. The same can apply to sludge incineration. Data on generation of waste from few European pulp and paper mills – Holmen, SCA, Norske Skog, Stora Enso are presented in Table 2.4 (Monte et al. 2009).

The amount of waste generated when virgin fibres are used as raw material depends mainly on the pulping process used (Table 2.5) (IPPC 2001; Monte et al. 2009). IPPC (2001) reports that in Europe, 65 % of total pulp production is kraft pulp which produces about 100 kg/Adt of wastes. Semi-chemical and mechanical processes produce about 60 kg/Adt.

CEPI (2006) has reported that in 2005, the total production of paper in Europe was 99.3 million tones. This generated 11 million tonnes of waste, representing about 11 % in relation to the total paper production. The production of recycled paper, during the same period, was 47.3 million tonnes generating 7.7 million tonnes of solid waste (about 70 % of total generated waste in papermaking) which represents 16 % of the total production from this raw material.

The amount of waste sludges generated from a mill using secondary fiber differ from a mill using virgin materials. Also, the composition is different. A greater amount of rejects is produced when processing recycled fiber, because of the unrecyclable filler proportion in the raw material. This problem is especially conspicuous in mills producing recycled paper from office waste, using highly filled grades as the raw material. Deinking mill sludge generally has a higher ash content; the kraft pulp mill sludge is found to be high on sulfur. Obviously, great variations occur within both plant types, depending on the processes and raw materials (Glenn 1997). The amount of wastes produced in paper mills based on recycled fibre depends mainly on the quality of recovered paper used as raw material. It also depends on the effort and expenses made in preparation of secondary fibres for certain product and process requirements. The average quantities of waste generated through production of different paper grades from recycled fibre are presented in Tables 2.6 and 2.7 (Kay 2002; Scott and Smith 1995; Gavrilescu 2008).

2.1 Generation of Wastewater Treatment Sludge

Joyce et al. (1979) have reported that about 40–50 kg of sludge (dry) is generated in the production of 1 tonne of paper at a paper mill in North America. Of that approximately 70 % is primary sludge and 30 % is secondary sludge (Elliot and Mahmood 2005, 2006). The primary sludge can be dewatered relatively easier. Compared with the primary sludge, the secondary sludge is very difficult to dewater. The secondary sludge consists mostly of excess biomass produced during the biological process (Ramalho 1983). About half of the incoming organic pollution load is converted into secondary sludge. The solid content is 0.5–2 % solids (Winkler 1993). Generally, a treatment plant includes both primary and secondary treatment stages installed one after another. Primary treatment stage is based on the sedimentation process mainly, but also can be implemented by a flotation method.

Table 2.6 Waste generated through production of different paper grades from recycled fibre

Paper grade	Solid waste (dry basis, kg/Adt)
Packaging paper	50–100
Newsprint	170–190
Light-weight coated paper/super-calendered paper	450–550
Tissue and market pulp	500–600

Based on Kay (2002)

Table 2.7 Rejects and sludge generation from different recovered paper grades and papers

Paper grade	Recovered paper grade	Total waste	Rejects		Sludges	
		Rejects and sludges	Heavy-mass and coarse	Light-mass and fine	Flotation de-inking	White water clarification
Market DIP	Office paper	32–46	<1	1 4–5	12–15	15–25
Graphic paper	News, magazines	15–20	1–2	3–5	8–13	3–5
	High grades	10–25	<1	≤3	7–16	1–5
Sanitary paper	News, magazines, office paper, medium grades	27–45	1–2	3–5	8–13	15–25
Liner, fluting	Old corrugated containers, Kraft papers	4–9	1–2	3–6	–	0–1
Board	Sorted mixed recovered paper, old corrugated containers	4–9	1–2	3–6	–	0–1

Based on Scott and Smith (1995), Gavrilescu (2008)

The secondary treatment stage, is based on biological treatment preformed in either aerobic lagoons, activated sludge systems, anaerobic treatment or sequential biological treatment (aerobic-anaerobic or anaerobic-aerobic) systems. Moreover, tertiary treatment can take place in addition to the above mentioned treatment stages in countries with tight environmental regulations (Bajpai 2000; Bahar 2009; Abubakr et al. 1995). About 80 % of total suspended solids contained in wastewater entering the treatment process are transferred to wastewater treatment sludge during the primary process (Monte et al. 2009). Inorganic part of wastewater treatment sludge is mostly present in the form of sand, while organic part is present as bark, fibre or other wood residuals. During the biological treatment, soluble organic materials are converted to carbon dioxide, water and biomass by microorganisms present in active sludge and required for successful process implementation. The excess biomass is settled in the secondary clarifier where secondary sludge also known as biological sludge, biosolids or activated sludge, is produced (Bahar 2009; Abubakr et al. 1995). Depending on a certain treatment scheme applied at a certain mill, primary sludge and secondary could be also either mixed together or collected separately. Basically, primary sludge consists of both organic and inorganic matter, while secondary wastewater treatment sludge consists mainly organic materials.

Secondary sludges are often difficult to handle (due to a high microbial protein content). It needs to be mixed with primary sludge to allow adequate dewatering prior to landfilling (McKeown 1979). Secondary sludges can be incinerated in existing boilers, but due to their low solids content, the steam generation capacity of a boiler is often reduced. This results in operational problems. Secondary sludges may be landfilled, but leaching of soluble nutrients may lead to the contamination of ground water (Saunarnaki 1988). Secondary sludges can be also applied to land as a soil improving organic fertiliser, as long as the material does not contain chlorinated organic compounds (or adsorbable organo-halogens), as most of these are acutely toxic to fauna and flora (Walden and Howard 1981; Saunamaki 1988; Bajpai et al. 1999). Chlorinated organic substances are present in the solid and liquid effluent of pulp and paper mills that use elemental chlorine or chlorine dioxide for bleaching of pulp (Bajpai et al. 1999; Gullichsen 1991).

Wastewater treatment plant residuals are presented by several types and their shares within all wastewater treatment plant residuals are presented in Fig. 2.1 (Bird and Talberth 2008; Boni et al. 2003). The figure shows that there is no certain tendency on how to collect sludge. Moreover, not all mills conduct biological wastewater treatment on-site. So, they do not generate secondary sludge at all. Depending on the method of sludge treatment and utilization, separation of different sludge types, especially secondary sludge, could be beneficial. For instance, if sludge is proposed to be used in the production of construction materials then the highest content of inorganic content is beneficial. Moreover, primary and secondary sludge types are of different nature and in the case of necessary pretreatment, different sludge should be treated separately to achieve best possible results.

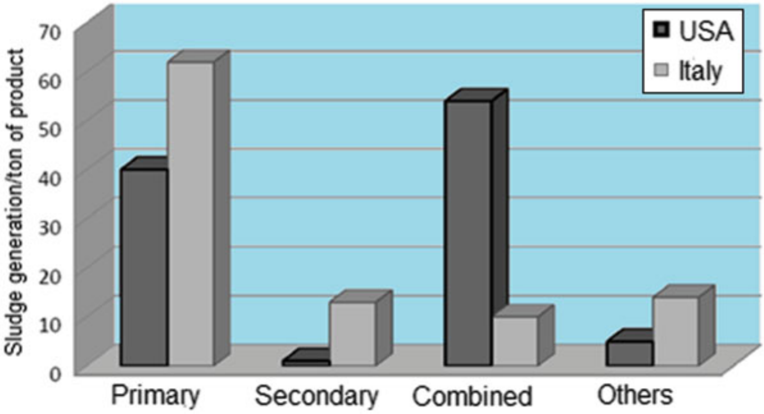

Fig. 2.1 Amount of different types of sludge generated by pulp and paper industry plants (Deviatkin 2013; Bird and Talberth 2008; Boni et al. 2003)

2.2 Generation of Deinking Sludge

Deinking sludge is generated in the mills producing recycled fibre from recycled paper (Bajpai 2006, 2013; Dash and Patel 1997; Seifert and Gilkey 1997). The amount of sludge on a dry mass basis can vary from 20 % in a newsprint mill to 40 % in a tissue mill. Deinking process enables increase of brightness and cleanliness of the material being produced so in many cases deinking process is included in the production scheme. Froth flotation deinking process is generally used in pulp and paper industry for selective deletion of ink particles only during recycled fibre processing. Wash deinking process is also used. This kind of deinking is aimed at removal of small particles, including fillers, coating materials, fines, and inks. However, froth flotation and wash deinking could be combined in the same production line so that the effect of unwanted materials removal is increased. In tissue paper production, in addition to deinking, de-ashing process is applied for better removal of fines and fillers (Kujala 2012; Deviatkin 2013). In case of deinking sludge, total suspended solids can be categorized into organic matter, such as bark and fiber, and inorganic matter, such as, kaolin, clay, calcium carbonate, titanium dioxide that are resulting from coating materials and other chemicals used for paper production.

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Chapter 3
Composition of Waste

Abstract The composition of waste generated in pulp and paper industry are presented. The composition of sludge depends on the raw material, manufacturing process, chemicals used, final products and the wastewater treatment technique. In case of recycled papers, it also depends on the type of paper used and the number and types of cleaning stages used in the recycling operation. The wastes are generated at different stages of the production process namely in the debarking, chipping, screening and cooking liquor clarification operations, in the maintenance of the plant and also in the treatment of fresh water and wastewater. The main solid by-products are lime-mud, green liquor sludge (dredge), recovery boiler ash, grits, bark, ashes and wastewater treatment sludge. If bark and other wood residues are not burned for energy recovery, they would represent the major fraction of the residues. Green liquor, lime mud and sludge mainly consist of calcium carbonate. Ash from the recovery boiler mainly consists of sodium sulphate and, to a lesser extent, sodium carbonate. Bark ash consists mainly of calcium oxide/ calcium carbonate and potassium salts. Grits mainly consist of calcium carbonate. Sludge from primary clarification consists primarily of fibres, fines and inorganic material in mills that employ fillers in their products. Sludge from biological treatment contains a high proportion of organic material. This sludge and the sludge from primary clarification represent the bulk of the wastes when using virgin fibres. Waste sludges from a mill using secondary fiber differ from a mill using virgin materials, not only by amount but also by composition.

Keywords Waste composition • Pulp and paper industry • Solid waste • Wastewater treatment technique • Lime-mud • Green liquor sludge • Dredge • Recovery boiler ash • Grits • Bark

3.1 Composition of Solid By-Streams

Kraft pulping process is the most common pulping process in the world (Smook 1992; Biermann 1996). As has been mentioned in the previous chapters, the main solid by-products are lime-mud, green liquor sludge (dredge), recovery boiler ash, grits, bark, ashes and wastewater treatment sludge. Bark and other wood residues would represent the major fraction of the residues if not burnt for energy recovery

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(Axegard and Backlund 2002; Demir et al. 2005; Martins et al. 2007). Green liquor, lime mud and sludge mainly consist of calcium carbonate. Ash from the recovery boiler mainly consists of sodium sulphate (approx. 85 %) and, to a lesser extent, sodium carbonate (15 %). Most of the recovery boiler fly ash and ESP (Electrostatic Precipitator) dust is returned to the chemical cycle. Approximately 5 kg/t of these residues is removed to control the sulphur balance. Bark ash consists mainly of calcium oxide/calcium carbonate and potassium salts. Grits mainly consist of calcium carbonate. Green liquor sludge, dredges and lime mud are often mixed and constitute the largest fraction of the overall solid waste generated in the kraft pulping process (Bajpai 2008). The composition in such a mixed waste varies widely, containing different amounts of metals as barium, chromium, copper, lead, nickel or zinc. The sulphite pulping production is related to the generation of different types of manufacturing-specific wastes, the majority of which can be further utilized (Smook 1992). The wastes are generated at different stages of the production process namely in the debarking, chipping, screening and cooking liquor clarification operations, in the maintenance of the plant and also in the treatment of fresh water and wastewater. The solid waste removed from mechanical pulping consists mainly of bark and wood residues from debarking, washing and screening of chips, primary sludge, ash from energy production and biological sludge. The main waste associated with this operation consists of different types of sludge, mainly fibre, containing primary sludge, and sludge from biological wastewater treatment. A chemical analysis of primary and secondary or biological sludge from a mechanical pulp mill is shown in Table 3.1 (IPPC 2001).

The waste generated in a paper mill producing paper from virgin pulp is small in comparison to production of recycled paper and pulp (Monte et al. 2009). Waste from paper production consists of rejects from stock preparation and sludge from water treatments. The rejects are normally led to the effluent treatment (IPPC 2001), but they may also be directly led to sludge dewatering. Most of the solids will end up in the primary sludge. Sludge from fresh water and wastewater treatments represents, in many mills, the main source of wastes. Following types of sludge are produced:

Table 3.1 Composition of primary sludge and biological sludge from mechanical pulp mill

Component	Primary sludge	Secondary sludge
Dry solid content (%)	48	32
Volatile solids (% DS)	33	48
TOC (%)	19	23
Copper (mg/kg DS)	238	71
Lead (mg/kg DS)	41	22
Cadmium (mg/kg DS)	<0,7	<0,7
Chromium (mg/kg DS)	24	17
Zinc (mg/kg DS)	141	135
Nickel (mg/kg DS)	6	8
Mercury (mg/kg DS)	0,1	0,09

Based on IPPC (2001)

- Sludge produced from chemical pre-treatment (chemical precipitation/flocculation) of surface water. This sludge is only produced in those mills using surface water with low quality. The amount of sludge can be significant in this case.
- Sludge from primary clarification is generated in most mills. This sludge consists mainly of fibres, fines and inorganic material in the mills using fillers in their products.
- Sludge generated during biological treatment contains a high proportion of organic material. This sludge and the sludge generated from primary clarification represent the bulk of the wastes when using virgin fibres.
- Sludge from chemical flocculation is produced in mills with tertiary effluent treatment. This treatment produces substantial amount of sludge. The amount of organic and inorganic material in the sludge varies from mill to mill; It depends on the dosage and type of flocculants used.

Most of the impurities present in recovered paper end up as waste (Blanco et al. 2004). The major waste materials are rejects, different types of sludges and ashes when the paper mill has an on-site waste incineration system. Depending on the grades of raw material, process design, manufactured product and wastewater treatment, different amounts and qualities of wastes are produced. The residues can be roughly subdivided in heavy and coarse rejects, light and fine rejects and sludge. Depending on its origin and nature, sludge again may itself be subdivided into deinking sludge, sludge from process water clarification in micro flotation units, and sludge from waste water treatment (primary sludge and secondary sludge from biological treatment). Rejects are impurities present in the recovered paper, consisting mainly of lumps of fibres, staples, metals from ring binders, sand, glass and plastics, and constitute about 6.5 % of the purchased recovered paper. Rejects are removed as much as possible in the earliest stage in the stock preparation section. The paper sludge generally contains very high levels of dry solids because it is rich in fibres and because of this it dewaters very easily.

The composition of sludge depends on: the raw material, manufacturing process, chemicals used, final products and the wastewater treatment technique. In case of recycled papers, it also depends on the type of paper used and the number and types of cleaning stages used in the recycling operation. Sludge from mixed office wastepaper may contain high levels of clay and other types of fillers, printing inks, stickies from envelope adhesives, as well as fibers and paper fines. Sludges from mixed office wastepaper recycling operations may contain as much as 2 % ash from fillers in the wastepaper. Sludge solids produced by pulp and paper mills typically include a majority fraction of fiber. Depending on the mill ink, sand, rock, biological solids, clay/fillers, boiler ash, grits from recausticizing, etc. may make up the other fractions. Because of the constituents that may exist, along with the water fraction, typical sludge analysis can vary widely. The amount of sludge generated per bale of raw material received varies by plant type. Kraft, sulfite and deinking mills by and large generate approximately 58, 102 and 234 kg of sludge per ton respectively (Scott and Smith 1995). These sludges contain approximately 40–50 % water by weight and a heating value of about 3,600 Btu/lb (dry) (Frederick et al. 1996).

Table 3.2 Ultimate analysis of different types of paper mill sludge

	Analysis (%)						
Sludge type	Solids	Ash	Carbon	Hydrogen	Sulphur	Oxygen	Nitrogen
Deinking mill 1	42.0	20.2	28.8	3.5	0.2	18.8	0.5
Deinking mill 2	42.0	14.0	31.1	4.4	0.2	30.1	0.9
Recycle mill	45.0	3.0	48.4	6.6	0.2	41.3	0.5
Bleached pulp mill	33.4	1.9	48.7	6.6	0.2	42.4	0.2
Pulp mill	42.0	4.9	51.6	5.7	0.9	29.3	0.9
Kraft mill 1	37.6	7.1	55.2	6.4	1.0	26.0	4.4
Kraft mill 2	40.0	8.0	48.0	5.7	0.8	36.3	1.2

Based on US EPA (2012)

Table 3.3 Macronutrients concentration in pulp and paper mill WWTP residues

Nutrient	Range	Median
Macronutrients (g/kg):		
Nitrogen (all mill types)	0.51–87.5	8.98
Nitrogen (combined mills)	1.1–59	8.5
Nitrogen (primary mills)	0.5 1–9.0	2.7
Nitrogen (secondary mills)	6.2–87.5	23.3
Phosphorous (all mill types)	0.01–25.4	2.35
Phosphorous (combined mills)	0.1–25.4	0.67
Phosphorous (primary mills)	0.01–4.0	1.6
Phosphorous (secondary mills)	0.42–16.7	4.2
Potassium	0.12–10	2.2
Calcium	0.28–2.10	14.0
Magnesium	0.2–19.0	1.55
Sulphur	0.2–20.0	4.68

Based on Bird and Talbert (2008), Thacker and Vriesman (1984)

The low heating value is a result of high levels of clay, calcium carbonate and titanium oxide. The ash content in paper mill sludge can be as high as 50 %. Not only do mills produce varying amounts of sludge, the sludges they produce are distinctly different in composition. High ash sludges have a significantly lower heating value than low ash sludges, which affect its suitability for certain disposal methods (e.g., incineration and gasification) (Scott and Smith 1995).

Table 3.2 shows ultimate analysis of different types of paper mill sludge (US EPA 2012) and Tables 3.3 and 3.4 show macronutrients and micronutrient concentration in pulp and paper mill WWTP residues (Bird and Talbert 2008; Thacker and Vriesman 1984; NCASI 1984).

For determining the best method for sludge disposal, it is very important to characterize the sludge carefully. Generally, sludge can be characterized by following parameters (Dahl 2008):

- Moisture content
- Ash content

Table 3.4 Micronutrient concentration in pulp and paper mill WWTP residues

Micronutrients (mg/Kg)		
B	<1–491	25.0
Cl	0.06–8,500	383
Cu	3.9–1,590	52.0
Fe	97.1–10,800	1,540
Mn	13–2,200	155.0
Mo	2.5–14.0	–
Zn	13–3,780	188

Based on Bird and Talbert (2008), Thacker and Vriesman (1984)

Table 3.5 Chemical composition of different types of sludge generated from various production processes

Source	Elements							Heating value on dry basis, MJ/kg
	Solids	Ash	Carbon	Hydrogen	Sulphur	Oxygen	Nitrogen	
(a) Abubakr et al. (1995)								
Kraft pulp mill	37.6	7.1	55.2	6.4	1.0	26.0	4.4	24.1
Pulp mill	42.0	4.9	51.6	5.7	0.9	29.3	0.9	21.5
Bleached pulp mill	33.4	1.9	48.7	6.6	0.2	42.4	0.2	20.1
Deinking sludge	42.0	20.2	28.8	3,5	0.2	18.8	0.5	12.0
Recycled paper mill	45.0	3,0	48.4	6.6	0.2	41.3	0.5	20.8
Bark	54.0	3.5	48.0	6.0	0.1	42.1	0.3	20.3
(b) Niessen (2002)								
Mixed sludge paper mill	–	9.6	45.9	6.5	0.7	9.6	3.7	18.9
Deinking sludge	–	50.1	26.9	2.9	0.2	18.8	1.2	8.6

- Heating value
- Loss of ignition
- Fiber length distribution
- Particle size distribution
- Viscosity
- pH

Chemical composition of different types of sludge from various production processes has been reported by several researchers (Gottsching 2000; Abubakr et al. 1995; Niessen 2002; Dahl 2008). Table 3.5 shows the data reported by Abubakr et al. (1995) and Niessen (2002). Deinking sludge reported by Niessen (2002) have the highest ash content. More than a half of the sludge is incombustible inorganic matter. Along with that, carbon content of the sludge is the lowest between all sludge types. The parameters mentioned directly affect heating value – the major characteristic of sludge as a fuel. Therefore, even if dry solids content of deinking

Table 3.6 Heavy metals content in different types of sludge

	Elements (dry basis)						
Source	Pb, mg/kg dry	Cd, mg/kg dry	Cr, mg/kg dry	Cu, mg/kg dry	Ni, mg/kg dry	Hg, mg/kg dry	Zn, mg/kg dry
Primary sludge	41	<0.7	24	238	6	0.1	141
Secondary sludge	22	<0.7	17	71	8	0.01	135
Deinking sludge from recovered paper	10–210	0.01–0.98	9–903	20–195	<10–31	0.1–0.9	34–1,320
Municipal solid waste	50–350	1–35	8–240	35–750	1–150	0.1–2	85–500

Based on Valkenburg et al. (2008), Monte et al. (2009)

Table 3.7 Ash analysis of deinking sludge

Ash analysis, weight %	Fe_2O	SiO_2	Al_2O_3	CaO	MnO	TiO_2	MnO	P_2O_5	Na_2O	K_2O	SO_3
Deinking sludge 1	2.2	28.6	43.4	4.6	0.2	2.8	0.2	5.4	3.6	1.3	4.1
Deinking sludge 2	1.3	40.9	22.9	25.8	6.9	1.8	–	–	0.2	0.2	–

Based on CANMET Energy technology centre (2005)

sludge is somewhat comparable to the rest sludge types or higher, heating values on dry basis are the lowest. The rest types of sludge have heating values comparable with bark. Table 3.6 shows heavy metals content in different types of sludge (Monte et al. 2009). Sludge contains smaller amounts of heavy metals compared to municipal solid waste. So, the flue gas treatment system suitable for municipal solid waste incineration plants can be easily applied to sludge incineration process to comply with the regulations set on heavy metals emissions. Table 3.7 shows composition of inorganic part of deinking sludge, i.e. ash left after incineration. Composition of ash can vary depending on the raw materials used for recycled fibre production. Inorganic part of deinking sludge mainly consists of Si, Al, and Ca oxides. The rest elements are present in smaller concentrations. Si and Al are the main components of kaolin, while calcium is the main element of precipitated calcium carbonate. These materials are widely used in paper production that is further used for manufacturing of recycled fibre. The composition determines the applicability of deinking sludge in the production of different types of construction materials. Some of the sludge properties, particularly its aggregate state, depend on dry solids content of sludge (Degremont 2007).

Comparing with other typical industrial sludges containing 16–35 % of total dry solids, the original secondary sludges from municipal wastewater treatment plant or a pulp and paper mill usually contains a much higher ratio of water (98–99 %) (Oral et al. 2005; Rato Nunes et al. 2008). A comparison of municipal and pulp and

Table 3.8 Comparison of municipal and pulp and paper activated sludge

	Municipal sludge	Pulp/paper activated sludge
Total dry solids (total solids) (%)	0.8–1.2	1.0–2.0
Volatile solids (% total solids)	59–68	65–97
Nitrogen (% total solids)	2.4–5.0	3.3–7.7
Phosphorous (% total solids)	0.5–0.7	0.5–2.8
Iron (g/kg_ total solids)	0	0.33–2.2
pH	6.5–8.0	6.0–7.6
Heating value (MJ/kg_ total solids)	19–23	22–25

Based on Elliott and Mahmood (2007)

paper secondary sludge characteristics is presented in Table 3.8. As can be seen that similarities exist between municipal and pulp and paper waste activated sludge.

Recyclers are producing two to four times more sludge as virgin pulp mills. Produced sludge can be considered to fall into two main types:

- High-ash sludge (>30 % dry weight)
- Low-ash sludge (<30 % dry weight)

High-ash sludges are chemical flocculation sludges generated by pulp mills, primary sludges generated by production of paper from recycled fibers and deinking sludges generated by paper mills, alternatively, low-ash sludge represents primary, secondary or biological sludges generated by pulp or paper mills (Table 3.9) (Méndez et al. 2009). Primary and deinking paper mill sludge constitutes a mixture of short cellulosic fibers and inorganic fillers, such as calcium carbonate, china clay, and residual chemicals dissolved in the water.

CEPI (2011) has reported the composition, energy content and composition of the dry matter content of different solid by-streams in European paper mills (Tables 3.10, 3.11, and 3.12).

Paper mill biosolids contain various plant nutrients including N, P, K, Ca and Mg. However, nutrient concentrations of paper mill biosolids vary according to the level of microbial decomposition that has occurred during secondary treatment (Vance 2000) and also the pulping method used. Nitrogen contents and carbon nitrogen ratio of paper mill biosolids vary widely depending on the type of paper being produced, the paper production process, and the type of raw material used. Primary biosolids consist of organic matter mainly in the form of cellulose (Jackson and Line 1997) or wood fiber settled out in a primary treatment and usually contain 0.3 % or less nitrogen by dry weight, with a carbon nitrogen ratio of more than 100:1 (Bellamy et al. 1995). In the deinking sludge, carbon content is consistently high while nitrogen and phosphorus contents are consistently low (NCASI 1991; Trépanier et al. 1996) which is similar to most wood residues like bark, sawdust, wood chips, etc. Secondary biosolids contain fibers and other fine materials that were not removed in the primary treatment. Bacteria decomposed the remaining organic matter that is contained in the water, i.e., the sugars and other constituents such as cellulose, and then they are collected through decantation of the treated water.

Table 3.9 Composition, heating values and cation exchange characteristics for different paper mill sludge

Paper mill sludge	Organic content (wt %)	Ash content (wt %)	Heat value (MJ/kg)	Cation exchange characteristics (cmol/kg)
High ash sludges				
Secondary sludge from paper mill producing paper from recycled cellulose without deinking process	67.23	32.77	16.5	17.30
Primary sludge from paper mill producing paper from recycled cellulose without deinking process	64.72	35.28	14.2	33.58
Deinking sludge from paper mill that recycled paper not recycled previously	60.36	39.64	12.0	18.12
Deinking sludge from paper mill producing newspaper	59.30	40.70	12.2	19.06
Low ash sludges				
Primary sludge from pulp mill producing pulp from virgin wood	94.31	5.69	20.1	30.20
Primary sludge from paper mill producing paper from virgin wood	93.79	6.21	19.8	32.41

Based on Méndez et al. (2009)

Table 3.10 Composition of sludges and rejects[a]

	Dry solids (%)	Organic matter[b] (%)	Mineral matter[c] (%)
Primary sludge	50	40	60
Secondary sludge	40–50	50	50
Deinking sludge	56	50	50
Coarse rejects	55	92	8
Screen rejects	55	90	10

Based on CEPI (2011)

[a] The composition can vary per paper mill. The figures in this table are an average based on different reports

[b] Of dry content

[c] Of dry content

Nitrogen, phosphorus and potassium fertilizers are added in the secondary treatment to allow microbial growth and activity. Therefore the secondary biosolids are very rich in nitrogen (3.0–4.0 %) and moderate in phosphorus (0.1–0.3 %) content (Bellamy et al. 1995; Zibliske 1987). Combined paper mill biosolids are prepared

Table 3.11 Energy contents of sludges and rejects

	Energy content (MJ/ton$_{\text{wet}}$)
Primary sludge	2,690
Secondary sludge	4,000–5,000
Deinking sludge	3,000
Coarse rejects	12,000
Screen rejects	8,000

Based on CEPI (2011)

Table 3.12 Composition of dry matter content of solid by-streams

	Content
Primary sludge	Fibres, fillers, coating clay, calcium carbonate
Secondary sludge	Calcium carbonate, cupper, micro organisms, fibres, proteins
Deinking sludge	Cellulose fibres, calcium carbonate, kaolin, ink
Coarse rejects	Recyclable fibres, wet strength fibres, plastics, wood, metal, others
Screen rejects	Cellulose, plastics, hair, stickies

Based on CEPI (2011)

by mixing primary and secondary biosolids in different proportions (50:50; 40:60 or 67:33) (Vance 2000). Combined paper mill biosolids usually contain 1.0–2.5 % nitrogen by dry weight. The carbon nitrogen ratio for these materials ranges from 100:1 to 20:1 and nitrogen phosphorous ratios for these materials ranges from 4:1 to 8:1 (Rashid et al. 2006). Nitrogen fertilizer is usually added to combined paper mill biosolids at the mill, before its delivery to farm sites and has high amounts of NH_4-nitrogen and NO_3-nitrogen contents (Bellamy et al. 1995).

About 150 chemicals can be detected in deinked paper mill biosolids (Beauchamp et al. 2002). In general the carbon, nitrogen, phosphorous and potassium contents of deinking paper mill biosolids are similar to those of primary paper mill biosolids. The contents of arsenic, boron, cadmium, cobalt, chromium, manganese, mercury, molybdenum, nickel, lead, selenium, and zinc are also low and showed low variability. However, the copper contents were above the Canadian compost regulation for unrestricted use and required a follow-up. The fatty acids and resin acids and polycyclic aromatic hydrocarbons were the organic chemicals measured at the highest concentrations. It was concluded that raw de-inking paper mill biosolids and its young compost do not represent a major threat for the environment but can require an environmental follow-up. Heavy metals in the sludge are known to pose potential health risks to plants and animals if present in too high concentrations. They are strongly retained by soils and therefore can persist for long periods in the environment. Heavy metals are present as contaminants in pulp mill sludge either as a result of chemicals added during the pulping process or they originate in the wood itself, having been adsorbed from soil by trees.

Cavka and Guo (2013) conducted compositional analysis of Fiber sludges from sulfate and sulfite processes. The analysis showed that sludges from sulfate process consisted mainly of glucan (69.1 %) and xylan (15.4 %) whereas sludges from

Table 3.13 Composition (% w/w) of sulfate and sulfite fiber sludges

Fiber sludge	Arabinan	Galactan	Glucan	Mannan	Xylan	Lignin	Ash
Sulfate fiber sludge	0.3	0.2	69.1	3.3	15.4	3.5	3.6
Sulfite fiber sludge	<0.02	0.1	89.7	2.7	1.6	0.8	1.7

Based on Cavka and Guo (2013)

sulfite process consisted mainly of glucan (89.7 %) and contained very low levels of other carbohydrates, such as mannan (2.7 %) and xylan (1.7 %). The content of lignin was low in both sulfate and sulfite processes (Table 3.13).

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Chapter 4
Pretreatment of Sludge

Abstract Pretreatment technologies of sludge – thickening, conditioning, dewatering, drying – are discussed. Thickening is a fundamental stage of sludge pretreatment. Pre-dewatering technologies include rotary sludge thickeners, gravity thickeners, dissolved air floatation clarifiers and belt presses. The most widespread thickening method is gravity thickening. Sludge from wastewater treatment plants is frequently conditioned, using chemical or physical means to alter the floc structures of the sludge imparting sufficient stiffness and incompressibility to the structures so that water entrained in the sludge can rapidly be drained through filtering or other means. Dewaterability of the sludge is very important because it determines the volume of waste that has to be handled. Dewatering of sludge is performed by using vacuum filters, belt filter presses, centrifuges, and membrane filter presses. Centrifuges and belt filter presses are currently the most popular dewatering methods due to their good operation and cost efficiency. The primary sludges can be dewatered easily as these are high in fiber and low in ash. The most difficult are the solids from the high-rate biological treatment systems. The primary sludge most difficult to dewater is that containing ground wood fines. Drying of sludge using flue gases from combustion process is a standard method. Fluidized bed dryers, rotary dryers, and multiple hearth dryers are found to be effective for sludge drying.

Keywords Sludge pretreatment • Thickening • Conditioning • Dewatering • Drying • Rotary sludge thickener • Gravity thickener • Dissolved air floatation • Belt press • Centrifuge

Sludge pretreatment consists of the following operations:

– Thickening
– Conditioning
– Dewatering
– Drying

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4.1 Thickening

Thickening is a essential stage of sludge pretreatment in any case (Diagileva et al. 2012). Pre-dewatering technologies which are commonly used include rotary sludge thickeners (RSTs) and gravity thickeners. Other technologies in use include gravity table thickeners, dissolved air floatation clarifiers and belt presses. The most well-known thickening method is gravity thickening. This does not involve installation of any complex equipment. Normally, gravity thickening is implemented in clarifiers. Once sludge has passed the clarifier, Dry Solids Content (DSC) increases from 0.3–3.0 to 7.0 %. Secondary sludge presented by excess biomass shows higher resistance to dewatering. For that reason, final DSC of secondary sludge cannot be more than 3.0 % in general (Hynninen 1998). The efficiency of sludge thickening is higher if thickening is done in a flotation unit, though the most efficient thickening method is that in a gravity table or in a belt thickener. The latter one allows increase of sludge DSC to as high as 15.0 %. Other technologies which are in use include, dissolved air floatation clarifiers and belt presses. Centrifuges, V-presses, coil vacuum filters and fabric vacuum filters are also used but the use of these is not very common. The floatation thickener used on secondary sludge can achieve approximately 4 % solids, whereas gravity thickener can achieve only about 2 % solids. Gravity thickener is a radial clarifier that is normally 11–12 m in diameter and 3–4 m in depth. The advantages of gravity thickeners include: simplicity, low operating costs, low operator attention and a degree of sludge storage. *Gravity thickening* reduces the water content of sludge. The energy consumption is also very low. Sludge is pumped directly to a circular tank which is equipped with a slowly rotating rake mechanism. This breaks the junction between the sludge particles and consequently increases the settling and compaction. With gravity thickening, the total sludge volume can be reduced by even 90 % from the original volume; this method consumes very little energy. Another target of gravity thickening is the significant hydraulic buffering capacity (up to 3 days) between the waste water stream and the sludge handling process. Gravity thickeners are being used in most of the large and medium-size waste water treatment plants. Conditioning chemicals are not normally required and there is minimal power consumption. However, these advantages are often offset by potential septicity/odour, less dewatering capability in comparison to other technologies and requirements of large space. These disadvantages have limited the use of gravity thickeners in recent installations. An RST is a rotary screen where water is removed by gravity and tumbling action. In many mills, RTSs have been installed before the screw presses as pre-dewatering units. With this type of pre-dewatering device depending upon the proportion of secondary sludge and the percentage of solids from the secondary and primary clarifiers, it is possible to increase the consistency to between 4 and 10 %. In a gravity table filter, sludge is drained on a rotary wire. Drainage is assisted by moving paddles. The paddles prevent the pluggage of wire. Gravity tables and RSTs produce sludges of similar consistency. Gravity tables are normally placed over screw presses to allow feeding by gravity. As with RSTs, polymers are used before the table filter. The performance of different types of pre-dewatering devices is presented in Table 4.1.

Table 4.1 Performance of pre-dewatering devices

Equipment	Expected solids (%)
Gravity thickeners	<3
Rotary sludge thickeners	4–10
Dissolved air floatation clarifiers	3–6
Gravity tables	4–10

Based on data from Kenny et al. (1997)

4.2 Conditioning

Conditioning is needed to make better water repulsion of sludge while being dewatered. Water-repellent properties of sludge are increased, since forms of water bonds are changed after conditioning. Thermal or reactant treatments are most commonly used to condition sludge at pulp and paper mills (Diagileva et al. 2012; Chen et al. 2002a, b). Reactant treatment that also refers to as chemical conditioning is used widely as an essential method. The best results are obtained when an inorganic salt and a polyelectrolyte are used together. Most frequently a combination of ferric chloride, aluminum oxides, and lime is used as an inorganic salt. Amount of chemicals used is determined by simple analysis. Reagents are fed into a flocculation tank for 1–3 min to allow reaction with sludge. As a result, small solid particles contained in sludge coagulate into larger ones that are simpler to dewater. Cost of reagents on local market determines mainly the method application. Moreover, about 60–70 % of used ferric chloride and 80–90 % of used calcium hydroxide pass into sludge, and therefore will influence composition of sludge generated (Degremont 2007; Hynninen 1998). Thermal treatment could be implemented by a wet air oxidation method or by heat treatment. Wet air oxidation method is flameless oxidation at temperatures of 230–290 °C and pressure of about 8.3 MPa, while the heat treatment method is used at temperatures of 180–200 °C and pressure level of 1–2 MPa. Heat treatment method is used more. When sludge is heated, water bound within the cell structure escapes from sludge and makes sludge easier to dewater. Sludge dewatering efficiency after thermal conditioning is higher, than that after chemical conditioning. However, cost of thermal conditioning is higher in comparison to chemical conditioning (Degremont 2007).

Sludge from wastewater treatment plants is commonly conditioned, using chemical or physical methods to alter the floc structures of the sludge. This is to impart sufficient stiffness and incompressibility to the structures so that water entrained in the sludge can rapidly be drained through filtering or by other means (Benitez et al. 1993; Wu et al. 1998). The functions of conditioners are as follows:

- Improve the sludge dewatering properties
- To reduce specific filtration resistance
- To enhance the dewatering efficiency

These increase the solids content after dewatering. There are four mechanisms through which chemical conditioners added to sludge at wastewater treatment plants act:

- Compression of the electrical double-layer
- Neutralization of charges
- Retention of precipitates
- Bridging effect

These actions destroy the stability of the existing flocs. This causes them to re-aggregate and precipitate into a tighter sludge filtration cake, thus enhancing water removal (Huang and Chang 1997). Generally lime, ferric chloride and polymers chemicals are used for conditioning, regardless of the type of equipment. These chemicals can be used separately or in combination. Ferric chloride is a very effective conditioning agent but suffers from disadvantage of being highly corrosive. The sludges that are difficult to dewater, require high dose of polymer. Wet air oxidation has also been used as a conditioning process to aid sludge dewatering. This process is commercially applied in the paper industry where filler recovery was a side benefit (Mertz and Jayne 1984). However, the brightness of the filler recovered was found to be lower than that of the filler grade clay and the installation was found to experience significant down time and high maintenance costs.

4.3 Dewatering

Dewaterability of the sludge is very important because it determines the volume of waste that has to be handled (Kerr 1997; Kantardjieff et al. 1997; Bajpai et al. 1999; Hewitt and Ellis 1983; Garg 2009; Diagileva et al. 2012; Degremont 2007; Hynninen 1998; Miller 2005; Navaee et al. 2006). If a sludge is dewatered from 1 % to 5 % solids, four-fifths of the volume of material has been eliminated, 10 % solids represents a nine-tenths volume reduction. The final volume of waste is directly related to the cost of disposal. The nature of sludge differs greatly with the type of pulp and paper manufactured. Table 4.2 presents the ranking of various sludges on their resistance to dewatering (Miner and Marshall 1976).

Vacuum filters, belt filter presses, centrifuges, and membrane filter presses are used for dewatering of sludge. Vacuum filters are among the earliest mechanical

Table 4.2 Relative dewaterability of pulp and paper mill sludge

Sludge	Ranking of resistance[a] to dewatering
Primary sludge (>20 % fiber or <30 % ash)	1
Primary sludge (>20 % fiber or >30 % ash)	2
Hydrous primary sludge (groundwood, glassine etc.)	3
Combined primary and secondary sludges	4
High rate treatment system biological solids	5
Post treatment alum or filter backwash solids	6

Based on data from Miner and Marshall (1976)

[a] Relative resistance to dewatering increases with number

devices used. Although many of its installations are being replaced by the more energy efficient belt filters, the use of vacuum filters is popular when used with precoat filtration. Another configuration is the rotary belt vacuum filter. The difference between a belt vacuum filter and a rotary drum vacuum filter is that the filter medium belt is wrapped around the surface of the drum rather than fixed to the drum. The advantages are continuous belt washing and more efficient discharge of cake (Santhanam et al. 1981). The operation of belt filter presses is quite similar to pressing in papermaking. The conditioned sludge is first drained under gravity and then sandwiched between two endless filtering belts. The pressure from the tensioned belts squeezes the water out of the sludge with a continuous cake discharged at about 15–25 % solid content when the feeding solid concentration varies from 2 to 5 %. Plate-and-frame filter presses can also be used if the volume of the sludge to be treated is not high. The rotary press is a recent development. Biosolids are pumped into a peripheral channel that has walls made of rotating filter elements. As the mechanism rotates, compression is created and as a result the liquid is forced through the filter elements. A cake is formed in the interior channel and then extruded (Kukenberger 1996). The belt filter press can be enhanced by acoustics or electrical osmotic dewatering techniques (Golla et al. 1992; Itawa et al. 1991; Hasatani 2001). Centrifugal dewatering uses the centrifugal force developed by spinning a bowl or basket to separate the sludge solids from the liquids. Disc, basket, and solid bowl centrifuges are all used for sludge dewatering with the latter being the most common. Solid bowl centrifuges are available in concurrent or counter-current flow designs (MacConnell et al. 1991). A typical centrifuge has two operating zones: a submerged pool and a drainage zone. The current generation of centrifuges can achieve solid contents of 25–35 % (Kukenberger 1996). Membrane filter presses are modified filter presses with the introduction of inflatable membrane systems, automatic cloth washing, controlled filling techniques, dual-speed plate separation and automatic discharge (Lowe and Shaw 1992). In some countries, the use of cross-flow micro-filtration technology has been used to concentrate sludge solids (Hudson and Lowe 1996).

The primary sludges can be dewatered easily as these are high in fiber and low in ash. The most difficult are the sludges from the high-rate biological treatment systems. The primary sludge most difficult to dewater is that containing ground wood fines. Primary sludges are normally tertiary or quaternary pulp and paper mill rejects, but often consist of quality fibers having a high monetary value. As the percentage of secondary sludge increases, the dewatering characteristics get worse, resulting in decreased cake solid contents. Tissue mills, NSSC plants, and recycle paperboard plants face problems with dewaterability of combined sludges. Sometimes, it may be advantageous to dewater the primary sludge separately from the secondary sludge. One example is a situation in which the secondary sludge can be disposed of through land application. Blended sludges are not usually suitable for such disposal. Another example is a situation in which the primary sludge can be used to produce a by-product or can be reused within the production process, but the blended sludge can not be used. If the combined consistency is less than 4–5 %, sludges must be pre-dewatered. It actually helps the dewatering process by reducing solution volume while increasing solid content for further dewatering, absorbing fluctuations of inlet solids consistency while stabilizing the output consistency, increasing outlet solids content and solids capture efficiency and reducing the overall consumption of polymer.

Centrifuges and belt filter presses are widely used dewatering methods due to their good operation and cost efficiency. Chamber filter presses are expensive compared to other presses. So these presses are used more in large applications elsewhere. Hydraulic presses, originally developed for the food industry with high hygienic demands, are also expensive. Screw presses are most suitable and used for sludges containing fibre material from the pulp and paper industry. The *decanter centrifuge* with its continuous feed and sludge output is the standard centrifuge type. High g (corresponding to the high multiples of the force of gravity, g) centrifuge models are favoured to achieve high dry solids content. The important elements are the bowl, which includes cylindrical and conical sections, the conveyor screw inside the bowl and the drive units to rotate them. The casing surrounding the bowl acts as a protective and noise suppression barrier, and channels the dewatered sludge cake and separated clarified liquid – or centrate – out from the unit. Centrifuges show higher efficiency with both inorganic and conditioned sludge and lower with organic sludge because of specific nature of secondary sludge. Final DSC after sludge processing in centrifuges typically varies from 10 to 35 % depending on sludge type and flocculants used for sludge conditioning. However, some advanced solid bowl decanters manufactured by Alfa Laval allow sludge dewatering up to 65 % DSC (Alfa Laval 2011). Specific energy consumption of a single centrifuge can vary from 20 to 60 kWh/tonne of sludge (Huber Technology 2013; CANMET 2005; Hynninen 1998). DSC of sludge processed in a belt filter-press varies in a range of 20–35 %. Energy needed for treatment of 1 tonne of suspended particles contained in sludge is 10–25 kWh (Degremont 2007). Screw presses show higher efficiency at primary sludge and DS treatment, while dewatering of secondary sludge is a questionable process because of slimy texture of secondary sludge. During normal operation of a screw press, final DSC of sludge can reach 30–50 % depending on a sludge type and a conditioning method applied. Specific energy consumption of a screw press is 10–30 kWh/tonne of sludge (Huber Technology 2013; CANMET 2005). With Chamber press, possible achievable DSC varies between 30 and 50 %. One of the modifications of chamber filter-presses is a membrane chamber filter-press, which efficiency of dewatering is as high as 70 % DSC at the end of the process. Drawbacks of the technology are its periodical work and high specific energy consumption of 30–90 kWh/tonne of sludge (Huber Technology 2013). Table 4.3 compares the performance of various dewatering devices and power consumptions.

With DAF (dissolved air flotation) clarifiers, secondary sludge is floated with dissolved air, usually with the help of some dewatering chemicals. Sludge is skimmed from the surface of the clarifier and the underflow re-treated in the aeration pond or the primary clarifier. In the DAF process, solids can be increased to 3–6 % for secondary sludges. The actual performance is frequently dependent upon the type of chemical applied and the dosage rate. DAF units also have the potential to eliminate odour problems. Few mills rely only on DAF units for sludge

Table 4.3 Comparison of performance and power consumptions

	Aerobic Sl.	Anaerobic Sl.	Power consumption	
Dewatering systems	% DS	% DS	kWh/t	kWh/(PT·a)
Gravity thickener	3–5	5–10	0–10	0–0.3
Belt filter or screw press	15–20	20–30	10–30	0.3–1.0
Simple decanter centrifuge	14–18	18–28	20–50	0.5–1.5
High-performance centrifuge	n/a	22–33	30–60	0.7–2.0
Frame and plate filter press	n/a	25–38	25–60	0.6–2.0
Membrane filter press	n/a	28–40	30–90	0.8–3.0

Based on Huber Technology (2013)

pre-dewatering (Kenny et al. 1997). Few activated sludge treatment plants use DAF units in combination with rotary sludge thickeners. One mill in Canada uses coil filters and V-presses to dewater primary sludge (Kenny et al. 1997). After pre-dewatering of the primary sludge, the secondary clarifier sludge and pre-dewatered primary sludge are mixed in a paddle mixer and then discharged for final dewatering on screw presses. No dewatering chemicals are required. Vacuum filter dewatering of biological sludges have been phased out of service in North America. Problems with poor capture rates, blinding and landfilling difficulties have eliminated this option.

During wastewater treatment, polymers are generally used for flocculation of sludge. Cyclodextrins (CDs) are reported to increase the performance of these polymers by increasing the cake solids and drainage rates of belt- or screw-pressed secondary or primary sludge (Banerjee 2009). These benefits are achieved at very low dosage of CDs. These are also found to decrease the specific resistance to filtration and increase the capture rate of solids during belt pressing. In three different full-scale trials, a combination of higher cake solids, better drainage, better filtrate clarity and lower polymer use was obtained. The use of CD for sludge dewatering has been implemented at the Mississippi mill. The results are found to be encouraging. About 30 % reduction in polymer costs for several months has been achieved. Several successful trials at other facilities in North America have been run successfully and additional implementations are expected. From a standpoint of cost, the CD is approximately twice the cost of the polymer. It displaces a much higher proportion of the polymer, so the cost benefits are attractive. The cost of α-CD is about three times higher than that of a typical sludge conditioning polymer, but since it is used at very low doses so the increase in overall chemical cost is relatively small. This cost is more than offset by the savings realized from the reduced polymer dosage. The benefits of the CD are incremental. CD basically boosts the performance of the polymer(s) applied with regard to cake solids, drying rate, and capture efficiency. The cost benefits are site-specific, but they are especially attractive at locations where sludge disposal costs are high. Finally, CDs are biologically derived products in that they are prepared from starch. Sludge conditioning polymers are derived from hydrocarbons, so that the displacement of polymers by CD carries both economic and socio-political benefit.

The application of nanosilica for paper mill dewatering has been studied by Taiwan researchers (Perng et al. 2006). The study was conducted in a paper mill in Taiwan which produces cultural and industrial paper products and uses sedimentation and a single-stage activated sludge process to treat its mill effluent. The primary sludge from sedimentation and the secondary sludge from the activated sludge stage were collected for this study. A conventional cationic polymer was utilized as a dewatering agent and a nano-silica preparation was used as co-agent to increase the dewatering efficiency. Sludge dewatering efficiencies were determined using the specific resistance to filtration and capillary suction time. A 23 factorial experimental design was used to delineate the effects and interactions of the sequence of polymer addition and the dosages. Analyses of the factorial design on the capillary suction time and specific resistance to filtration tests revealed that both the primary sludge and secondary sludge had similar treatment behaviors. All three variables under investigation were significant, but none showed interactions with each other. The secondary sludge had a poorer dewatering efficiency than did the primary sludge on the capillary suction time and specific resistance to filtration tests. The researchers found that the cationic polymer should be added first, followed by the anionic nano-silica. The reverse sequence of addition was largely harmful to the dewatering of the primary sludge. Both the cationic polymer and nano-silica showed close weighting factors on the dewatering efficiency.

In some cases, the primary dewatering device – rotary vacuum filters, centrifuges, V presses, twin-wire presses and screw presses – is followed by a press in order to further increase the solids content. The most popular device is vacuum filter. Solids capture in vacuum filter is 90–95 %, and the cake produced contain about 20 % solids. In order for the filter cake to discharge properly from the filter, 10–20 % long fiber ($>$100 mesh) must be present in the sludge (Miner and Marshall 1976). Vacuum filter cakes containing combined sludge solids can be further dewatered on V-presses to approximately 35–40 % consistency. A V press is just two discs providing a converging nip that applies pressure to the sludge to squeeze out the water. Vacuum filters can be equipped with either fabric media or steel coils. Fabric media are often used in situations when fiber content is low, the ash content is high, or the solids are otherwise difficult to dewater on a coil filter. The power costs for operating the large vacuum pump required by a vacuum filter are quite high. Nowadays vacuum filters are being replaced by belt presses, which seem to perform as well if not better, at lower operating cost.

Voith Paper has developed Thune which is a new design of screw press used for dewatering pulp and paper mill sludge (Norli and Smedsrud 2006). The trial was conducted at the new Adolf Jass Schwarza mill at Rudolstadt Germany in 2005. The new screw press achieves high torque which is distributed equally along the axis by integrating the inlet and discharge housings and the screen supports into the machine frame. The centre line of the press is kept low in order to minimise deflection at high loadings, the height above mountings being only 270 mm. The operating cost is kept low and the machine has been designed to facilitate maintenance and servicing. This new press achieves a higher dewatering per screen area than comparable sludge presses. The Thune SPS70 screw press at Schwarza handles all fine and sludge for dewatering, fed by a Meri BlueDrain gravity table. A Meri Sediphant is used to pre-dewater cleaner reject and pre-screened sewer matter. Dynamic torque control ensures a uniform consistency of the discharge. Voith Paper dewatering centre at Tranby Norway have also installed a smaller system at Orbro Kartong in Sweden, with a Meri Elephant filter and a Thune screw press.

Screw presses for sludge dewatering have become an accepted and well-proven alternative for decanters. HUBER RoS 3Q screw presses have previously been used mainly on wastewater treatment plants smaller than 20,000 PE. Recently, however, also more and more operators of bigger plants have become aware of the advantages screw presses offer in terms of operating reliability, dewatering results and energy efficiency.

HUBER could win the contract for the supply of a sludge dewatering plant for a new 140,000 PE wastewater treatment plant to be built on the Georgian Black Sea Coast where statically thickened and aerobic stabilised (10 m^3/h each) with 3 % solids content should be dewatered to at least 20 % to reduce sludge volume prior to solar drying (Huber Technology 2013). Measurements during plant start-up showed that the sludge had a solids content of only 0.7–1 %, i.e. it was not statically dewatered. The results achieved by the screw presses were therefore all the more surprising: Dewatering degrees of 29–33 % were achieved right from the beginning with a throughput of 15 m^3/h. The local operators optimised polymer dosing and could thus reduce coagulant agent consumption from initially 12–8 kg/t DR. Even with these low polymer doses the dewatering degree measured was still continuously 33 %. The screw presses reduce sludge volume prior to solar drying by 97 %. Even with this extraordinary sludge volume reduction power consumption of the screw press unit lies still below 1 kW. Another installation with two RoS 3Q 800 units in the Northeast of the USA achieves also dewatering degrees in excess of 30 % with an inlet solids content of only 1 %.

Disc centrifuges have found little application in the paper industry. They have been tried as thickening devices but experience has not been satisfactory. Basket centrifuges have been used to a limited extent for sludges that are very difficult to dewater. These centrifuges operate in a batch mode rather than continuously. Usually it is desirable to use the continuous decanter scroll centrifuge. Special scroll units have been developed for secondary sludge, and they are usually preferred over the basket centrifuge. Scroll centrifuges dewatering combined paper industry sludges generally produce cakes of 20–40 % consistency at solids capture efficiencies of 85–98 % from sludges conditioned with polymer. As the centrifuges operate on the basis of density difference separation, the sludges which are much denser than water, such as high-ash sludges, provide the best application of centrifuges. Specially designed scroll centrifuges can dewater secondary sludge from 2 to 11 % solids with 99.9 % capture efficiency (Reilly and Krepps 1982). However, it required 6–8 kg polymer per ton of sludge for conditioning. Centrifuges have a relatively low capital cost but can be expensive to operate due to requirement of chemical conditioning agents, their high power requirements, and their maintenance costs. Dissatisfaction with centrifugation has been attributed to the following:

- Generation of poor quality supernatant that could cause a buildup of fines in the treatment system,
- Susceptibility of centrifuges to plugging with pieces of bark
- The severe screw conveyor abrasion experienced at many mills

V presses have been used with success to the dewatering centrifuge and vacuum filter cakes containing as much as 30 % biological solids. However, the combined sludges normally encountered require sufficient conditioning for vacuum filtration or centrifugation to render them amenable to V pressing (Miner and Marshall 1976). V presses can be used to increase the solids content of the sludge high enough for incineration (Stoval and Berry 1969). V presses generally produce primary sludge cake consistencies of 30–45 %. Either a V press or a screw press would precede most bark boilers burning bark and sludge. The sludge would enter the press at 15–25 % solids and be subjected to a pressure of 690 kPa to increase the solids content to 30–45 % suitable for incineration (McKeown 1979).

The most powerful dewatering devices available are pressure filters. For combined sludge, cake of 30–35 % consistency can be produced with solids capture efficiency of 95–100 % (Miner and Marshall 1976). But, it is very important to precoat the filter cloth to facilitate cake discharge and reduce the frequency of media cleaning. Precoating is done with diatomaceous earth, flyash, cement dust etc. Media cleanliness has been indicated as a crucial parameter in determining the pressure filter cycle time. Pressure filtration also requires conditioning of the sludge before filtration. On pure secondary sludge, 35–40 % cake solids can be achieved with a conditioning agent and a pressure of 200–250 psi. The main drawback of the pressure filter is that it is a batch operation and requires a lot of operator attention. Continuously operating automatic units have also been developed, but they are mechanically complex and therefore subject to many maintenance problems.

Many paper mills have installed moving-belt presses (Twin-wire press). These presses have received intensive industry interest in the past. Moving-belt presses have generated cakes of a consistency comparable to that of two-stage dewatering with V presses, and with similar or somewhat higher conditioning costs and generally lower power consumption on primary or combined sludges. Polymers are generally used for the sludge conditioning, and some processes use dual-polymer systems. The cake solids are 20–50 % for the primary sludge whereas they are 10–20 % solids for the secondary sludge. Capture efficiency is found to be very high for belt presses, about 95–99 % of the solids fed. Requirement of the operator attention is low. These presses are energy efficient as they require power only to drive the belt. Another benefit is their ability to operate on secondary biological sludge. However, the major operating problem is belt life, which is only few months. The usual cause of failure is puncture of the belt by incompressible objects in the sludge. Also the press is subjected to corrosion due to hydrogen sulfide gas that is sometimes produced if there is any sulfur content in the sludge.

Toole and Kirkland (1984) reported screw press of new design for sludge-dewatering. These presses produce cake solids of 50–55 % when operated as the only sludge dewatering device. Solids capture ranges from 70 to 88 % with no polymer addition on primary sludge. Biological solids adversely affect solids recovery. Polymer can be used to improve efficiency but it has little or no effect on final sludge consistency; therefore is often not used on primary sludge. With secondary sludge, polymer is used. These presses appear to be energy efficient. Screw presses are replacing twin-wire presses as the dewatering technology of choice for the pulp and paper industry.

Zawadzki et al. (1997) developed an innovative and energy efficient sludge dewatering technology. The technology, impulse drying, involves briefly contacting the sludge under pressure with a heated surface. A unique feature of impulse drying is that the majority of moisture removal occurs as lipid water. A pilot- and laboratory-scale demonstration was conducted on low ash primary paper mill sludge. The pilot-scale impulse dryer was able to increase the sludge solids level 23.2 % greater than belt pressing alone. Study shows that impulse drying is effective on a wide variety of sludge types: primary (high and low ash), secondary, and mixed sludges. Impulse drying efficiency depends upon the presence of sufficient moisture in the sludge. Preliminary results indicate that the process is effective for sludges with initial solids content in the range of 15–50 %. Since, a sludge solids content of 30 % is typical of the performance of an inexpensive belt press, these experiments demonstrate the potential of retrofitting existing belt presses with an add-on impulse dryer. A high percentage of liquid water removal is achieved by the impulse drying process, which gives the technology an economic advantage relative to thermal drying. The liquid water removal has major cost reduction potential for either sludge burning or landfilling.

Technical Research Centre of Finland, VTT has studied improvement of sludge dewatering ability using ultrasound. Sludge contains voluminous microbes which contain much intracellular water which can be released by power ultrasound. About 30 % of water bound in sludge can be liberated to aqueous phase which is easy to remove in mechanical dewatering (www.greennetfinland.fi/fi/images/4/4b/Nordic_sludge.pdf).

Andritz has recently supplied Stora Enso with a new sludge dewatering line for thickening and dewatering of fibrous sludges (a gravity table and a sludge screw press to process up to 50 tonnes of mixed sludge per day) at the company's Skoghall Mill in Sweden (Andritz, 2013). The Skoghall Mill is an integrated pulp and paperboard mill. The mill has the capacity to produce over 700,000 tonnes per year of paperboard used for packaging liquids. The material to be dewatered consists of a mixture of fibrous and biological sludges, as well as chemical sludges from the mill's CTMP (chemical-thermomechanical pulp) line. Due to the higher final dryness achieved through the dewatering lines, Stora Enso Skoghall will be able to feed a higher amount of dewatered sludge to the power boiler and operate it with greater energy efficiency.

Investment costs for sludge dewatering in a 1,500 ADt/d newsprint mill are as follows:

Wire press	1.5–1.8 MEuros
Screw press	1.7–2.0 MEuros
Centrifuge	0.7–0.9 MEuros

The operating costs, assuming dewatering of both primary and biological sludge, are 0.3–0.6 MEuros/a. The cost is very dependent on the sludge composition and demand of flocculation chemicals (European Commission 2001).

4.4 Drying

Moisture content of dewatered sludge can be appropriate for sludge disposal, but not for energy or material recovery (Deviatkin 2013). Therefore, drying stage could be a important step in sludge pretreatment prior to its energy or material recovery. Drying of sludge using flue gases from combustion process is a standard method. This can be done either at sludge generating mill premises, or at a receiving plant, particularly, in sludge incineration plants. Fluidized bed dryers, rotary dryers, and multiple hearth dryers are found to be effective for sludge drying. In the rotary dryer, the sludge to be dried and air for drying are fed from the same end of the dryer, while air passes through a heater installed in the dryer. The inclined dryer slowly rotates moving sludge towards the discharge end of the dryer and the sludge is dried through the warm air stream. Rotary dryer could be either with direct or indirect contact of sludge and drying agent. If sludge drying is implemented in a rotary dryer with direct contact, then both sludge and drying agent are fed inside the dryer. In that case, hot exhaust gases are applied. When sludge is dried in a dryer with indirect contact, then drying agent is fed into a drum jacket, thus, preventing contact between sludge and the drying agent. In general terms, indirect dryers are better suited for drying of particles with low size and density and, therefore, rotary dryers with direct contact are more suitable for deinking sludge drying (CANMET 2005). Fluidized bed dryers allow implementation of fast and even sludge drying at low temperatures enabling utilization of waste heat, such as low-pressure steam. In the fluidized bed dryer the hot air coming from the bottom of the dryer through a straightening vane is used for heating up inert material, like sand, that later will contact with sludge enabling its drying. After sludge particles have been dried, they become lighter and overflow to a cooler. The most beneficial side of fluidized bed dryers is that almost all surface of sludge particles stays in contact with the drying agent (CANMET 2005). Creation of multiple hearth dryers was caused by the need of sewage sludge drying. Nonetheless, area of multiple hearth dryers application was extended to Pulp and Paper mills since not much difference can be found in sludge properties important for drying (Deviatkin 2013). Long residence time of sludge in the dryer allows effective drying of sludge by hot air blown inside. With the furnace type development, a modification allowing combination of drying and incineration processes became more popular.

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Chapter 5
New Technologies for Energy Recovery from Waste

Abstract New Technologies – Pyrolysis, Direct liquefaction, Wet Air Oxidation, Super Critical Water Oxidation, Steam Reforming, Gasification (Plasma Gasification, Super Critical gasification) for energy recovery from waste are discussed. The operating conditions (temperature, pressure, atmosphere and products, etc.) vary among the methods. For example, gasification and SCWO methods utilize air or oxygen while some methods are conducted under oxygen depleted or anaerobic conditions. Pyrolysis and gasification operate at high temperatures; Pyrolysis targets a high yield of oil, and gasification favors production of gas. The greatest sludge volume reduction (over 90 %) can be achieved with the high-temperature methods which is advantageous as it effectively reduces the physical amount of sludge for disposal. The major disadvantage for these high-temperature processes is their lower net energy efficiency for the treatment of secondary sludge containing very high content of water, resulting from the need of the energy intensive operations of dewatering/thickening and complete evaporation of the water in the sludge. In contrast, the other treatment methods, i.e., direct liquefaction, SCWO operate at a relatively lower temperature and more importantly without the need of dewatering /thickening and complete evaporation of the water in the sludge. Accordingly, these methods are more promising for the treatment of secondary sludge from the standpoint of energy recovery.

Keywords Energy recovery • Waste • Pyrolysis • Direct liquefaction • Wet air oxidation • Super critical water oxidation • Steam reforming • Plasma gasification • Super critical gasification

5.1 Pyrolysis

Pyrolysis process is also called destructive distillation. In this process, indirect heat is applied and the volatiles are captured. This technology is an alternative to incineration and landfill of paper mill wastewater sludge, though it generally requires a consistent waste stream to produce a usable fuel product (Monte et al. 2009; Fio Rito 1993; Frederik et al. 1996; Kay 2002, 2003; Fytili and Zabaniotou 2008). Xu and Lancaster (2008, 2009) have provided comprehensive information on pyrolysis of secondary sludge from pulp and paper industry and Lou et al. (2012)

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have provided information on pyrolysis of deinking sludge. The organic material is heated up to 400–800 °C in anaerobic conditions. A mixture of solid charcoal, water, water-soluble organics (pyroligneous acids, including methanol and acetic acid); water-insoluble organics grouped under the term of "tar" or "bio-oil", and non-condensable gases (hydrogen, methane, carbon monooxide, carbon dioxide) are produced. Long exposure times are required to optimize the production of char (CANMET 2005). No oxygen is allowed to enter the retort during the decomposition operation. Therefore, no combustion can take place. This technology has been developed for wastes with a high carbon content, such as wood, petroleum and plastic wastes.

Operational temperature, heating rate and reaction time are optimized in order to select the product being produced. If the process is operated at low temperatures and low heating rates then char is produced mostly. The char has high heating value, if sludge is mostly organic, and can be used as a fuel, or can be activated to become activated carbon. In contrast, high oil generation rate is gained at high heating rate at a temperature of 500 °C and higher and short residence time of sludge. As a result of such conditions, yield of approximately 70–75 % of bio-oil with lower heating value of 16–19 MJ/kg can be reached. In addition, solid material left after deinking sludge pyrolysis can be recycled in paper production process (Lou et al. 2012). Fast pyrolysis processes are implemented in fluidized bed boiler and circulating fluidized bed boiler or rotating cone reactors (Xu and Lancaster 2012). To summarize, pyrolysis is the process enabling not only energy recovery, but also material recovery. Energy recovery is implemented through produced bio-oil utilization, and material recovery is achieved by recovering of solids left after the process. However, important issue is the requirements set on dry solids content of sludge that should be more than 80 %. The commonly used reactors for fast pyrolysis include (Mohan et al. 2006):

- Bubbling fluidized bed
- Circulating fluidized bed
- Ablative
- Entrained flow
- Rotating cone reactors
- Vacuum reactors

In the pyrolysis process, the carbohydrate polymers – cellulose and hemicellulose – are degraded and converted. By proper selection of the operating conditions of temperature, heating rate and reaction time, pyrolysis process can be optimised to produce char, oil and/or gas. When heated at a temperature higher than 300 °C, the carbohydrate polymers de-polymerize into short chains of sugars which is accompanied by slow dehydration and subsequent reactions to form unsaturated polymer intermediates that may be eventually condensed to form char (Lomax et al. 1991). When heated at a higher heating rate to a higher temperature, the de-polymerization reactions will liberate volatile products as oil or tar. Cleavage of carbon-carbon bond will occur at a high temperature, leading to formation of gas products. If the objective is to maximize the oil yield then a high heating rate

Table 5.1 Typical properties of bio-oil and of a heavy fuel oil

Physical property	Pyrolysis bio-oil	Petroleum-based heavy fuel oil
Moisture content (wt %)	15–30	0.1
Specific gravity	1.2	0.94
pH	2.5	–
Elemental composition (wt %)		
Carbon	54–58	85
Hydrogen	5.5–7.0	11
Oxygen	35–40	1.0
Nitrogen	0–0.2	0.3

Based on Czernik and Bridgewater (2004)

and short gas residence time would be required, while for high char production, a low temperature and low heating rate would be preferred. High yield of bio-oil up to about 70–75 % can be produced in fast pyrolysis processes with very short residence time and elevated reactor temperature of ~500 °C or higher (Agblevor et al. 1995). A significant amount of char and equal amounts of oil and gas products can be obtained in slow pyrolysis processes operating at a low temperature for a long residence time. The char produced typically has a higher heating value of ~30 MJ/kg, which can be used as a valuable fuel for generating heat and electricity, or can be turned into activated carbon by activation. Pyrolysis oils are normally composed of a variety of organic oxygenates and polymeric carbohydrate and lignin fragments. These are derived from the thermal cracking of cellulose, hemi-cellulose and lignin components of the biomass (Mohan et al. 2006; Tsai et al. 2007). The comparison of physical properties of wood fast-pyrolysis oil are compared with those of a petroleum-based heavy fuel oil and are presented in Table 5.1 (Czernik and Bridgewater 2004). Pyrolysis oil contains a high concentration of water (15–30 %), and is highly acidic (corrosive) and unstable liquid. It has a lower caloric value of 16–19 MJ/kg in comparison with 40 MJ/kg for the petroleum-based heavy oil (Table 5.1). Pyrolysis oil can be used as a substitute for fossil fuels to generate heat, power and/or chemicals. Short-term applications are boilers and furnaces including power stations, whereas turbines and diesel engines may become available on the somewhat longer term. Upgrading of the pyrolysis oil to a transportation fuel is technically feasible, but needs further development. Transportation fuels such as methanol and Fischer-Tropsch fuels can be derived from the pyrolysis oil through synthesis gas processes. Furthermore, there is a wide range of chemicals that can be extracted or derived from the pyrolysis oil (Vitolo et al. 1999). Its energy density is four to five times higher than the original solid material, which offers important logistic advantages.

Pyrolysis is found to be more advantageous over conventional incineration processes for the treatment of sewage-sludge with respect to fuel economy, energy recovery, and the control of heavy-metal emissions (Lewis 1975). However, process efficiency is affected by sludge moisture content, such that co-pyrolysis with other wastes has been recommended in order to increase the dry-solids content of the sludge (Olexseyr 1975). The process normally needs additional treatment to remove excess water. Feedstocks containing higher water content cause increase in the production of hydrogen and methane, but these do not compensate for the losses of carbon monoxide and thermal efficiency (Carre et al. 1989). Water reduction is performed by dewatering to about 25 % dry solids followed by thermal drying to 95 % dry solids.

Bridle and Hertle (1988) reported an oil-from-sludge (OFS) process. This is an extension of standard pyrolysis process, with the system arranged to maximise the production of high quality oil, which can be used as a fuel. In this process, pre-dried sludge having 25 % dry solids is heated to 450 °C for a heating period of about 30 min under anoxic conditions; the pressure is kept just above atmospheric, until about 50 % of the sludge is evaporated. The vapours generated are then contacted with residual tar to catalyze the formation of high caloric value hydrocarbons. This process can produce 200–300 L of oil per tonne of dried sludge. In comparison with incineration and anaerobic digestion, 95–98 % of the energy in the dried sludge is recovered in the various products, and the net energy efficiency could be greater. However, the energy input for pyrolysis, including energy consumption for thickening, drying, and heating of the sludge feedstock to necessitate the pyrolysis process, is still very much high.

5.2 Direct Liquefaction

Extensive research on direct liquefaction has been conducted in the 1980s for the purpose of alternative energy production (Kranich 1984; Beckman and Elliot 1985; Boocock and Sherman 1985). Liquefaction can be accomplished indirectly or directly:

- For indirect liquefaction, biomass is converted into liquid products through first gasification to syngas followed by catalytic conversion (Dry 1999).
- Direct liquefaction of biomass feedstocks into liquid oils has attracted more intensive interest, due to its simpler technical route and better conversion economy and efficiency relative to the indirect liquefaction processes. In a typical direct liquefaction process, biomass is converted to liquid products directly but through a complex sequence of processes involving:

  Solvolysis
  Depolymerization
  Decarboxylation
  Dehydration
  Hydrogenolysis/hydrogenation (when hydrogen is present in liquefaction)

Direct liquefaction is a low-temperature, high-pressure conversion in the liquid phase, usually with a high hydrogen partial pressure and a catalyst to enhance the rate of reaction (Furness et al. 2000). Yields of the products depend on several parameters:

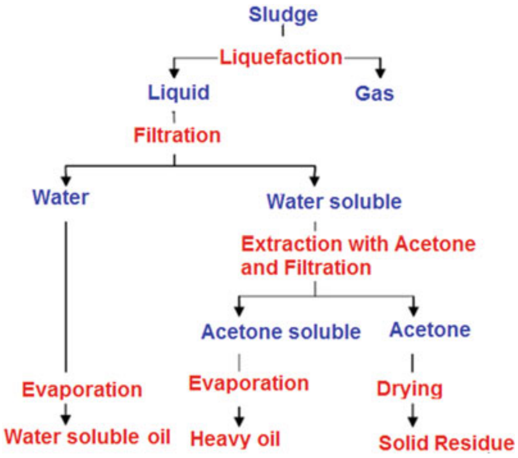


Fig. 5.1 Direct liquefaction process for sludge treatment (Based on Xu and Lancestor 2008, 2009)

- Temperature,
- Residence time,
- Initial biomass concentration,
- Catalysts
- Liquefaction atmosphere (inert nitrogen or reducing hydrogen)

The effects on the liquefaction product yields have been investigated. The process product is bio-oil as that of the pyrolysis. The reason to apply direct liquefaction process instead of pyrolysis is low quality of the product gas produced in the pyrolysis. Further processes are shown in Fig. 5.1 (Xu and Lancaster 2008, 2009).

In the study by Xu and Lancaster (2008), sludge treatment was conducted at temperature of 250–380 °C and 15–120 min residence time in the atmosphere of nitrogen or hydrogen. In this study sludge was absolutely dried before treatment and that implies huge energy expenses in the industrial scale application. The process was proven as energy deficient and requires further development. Co-liquefaction of secondary pulp/paper-mill sludge (solids concentration: 1.6 wt%) and waste newspaper with a total solids concentration of 11.3 wt% was investigated with and without the addition of catalysts in a 75 ml Parr High-Pressure reactor at temperatures of 250–380 °C for 20 min. The yield of heavy oil without catalyst was between 16.7 and 28.0 wt% within this temperature range, and reached maximum at 350 °C. The addition of formic acid, iron sulphide, or potassium hydroxide at 5 wt% of the total solids (on a dry basis) was found to enhance the heavy oil yield at 300 °C, particularly formic acid, which increased the yield of heavy oil from 24.9 to 34.4 wt%. Synergistic effects were observed between secondary pulp/paper-mill sludge and waste newspaper in the co-liquefaction operations. For example, the heavy oil yield attained was 26.9 wt% at 300 °C in the co-liquefaction of the mixture of 33 wt% sludge and 67 wt% waste newspaper, and was found to be 9 wt% and 6 wt% higher than the yields obtained from liquefaction of sludge and waste newspaper alone, respectively. The heavy oils from liquefaction or co-liquefaction at 300 °C for 20 min exhibited significantly higher energy contents (HHV ≥30 MJ/kg), almost doubled those (˜16 MJ/kg) of the original feedstocks. The work demonstrated that secondary pulp/paper sludge powder, with a higher heating value of 18.3 MJ/kg on a dry basis, could be effectively converted into liquid oil products by direct liquefaction in hot-compressed water with and without catalyst. Treatments of secondary pulp/paper sludge in water at 250–380 °C for 15–120 min in the presence of nitrogen atmosphere resulted in yields of water-soluble oils at 20–45 wt% and yields of heavy oils at 15–25 wt%, with higher heating values of 10–15 and >35 MJ/kg, respectively. The higher caloric values for the heavy oil products were accounted for by their compositions of long-chain carboxylic acids, heterocyclic nitrogen compounds and phenolic compounds and derivatives as evidenced by the gas chromatograph (GCMS) measurements. The liquefaction product yields were significantly influenced by the liquefaction temperature, the residence time, the initial biomass concentration, catalysts and the liquefaction atmosphere (inert or reducing). Within the temperature range (250–380 °C) tested, the lowest temperature produced the highest yield of total oils (at 60 wt%), while the greatest yield of heavy oil (at about 24 wt%) was obtained at 350 °C. If the temperature was fixed at 280 °C, a greater yield of heavy oil (reaching as high as 25 wt% for 120 min) was obtained as the length of reaction time increased. Similarly, a higher initial biomass concentration produced a greater yield of heavy oil but a reduced yield of water-soluble oil. The presence of 0.1 M potassium carbonate dramatically enhanced organic conversion, but suppressed the formation of both heavy oil and water-soluble oil. The use of the two alkaline earth metal catalysts, i.e., calcium hydroxide and barium hydroxide, did not alter organic conversion, but it catalyzed the formation of water-soluble oil and produced higher yields of total oil products. It was also demonstrated that the reducing atmosphere (i.e., hydrogen) in the liquefaction process promoted the heavy oil formation while suppressing the water-soluble oil formation. With the presence of 0.1 M calcium hydroxide and 2 MPa hydrogen, liquefaction of the sludge powder in water at 280 °C for 60 min produced a higher yield of heavy oil (26 wt%), almost two times as high as that in nitrogen (13.6 wt%), resulting in a greater net energy efficiency. It was thus suggested that direct liquefaction of secondary pulp/paper sludge in hot-compressed water with calcium hydroxide catalyst and in the presence of hydrogen could be an effective approach to recovering energy from the waste for production of liquid oil products.

5.3 Wet Air Oxidation

Wet oxidation or wet air oxidation (WAO) process can be defined as "the oxidation of organic and inorganic substances in an aqueous solution or suspension by means of oxygen or air at elevated temperatures and pressures either in the presence or absence of catalysts" (Zimmerman and Diddams 1960; Zou et al. 2007). The technology was first commercialized for the production of artificial vanilla flavoring and later for the treatment of paper-mill sludge and biological sludge. The application of this technology has expanded these days most successfully for treatment of industrial wastes such as the caustic solution from scrubbing towers, and for treatment of powdered activated carbon (Maugans and Ellis 2004). Other applications include the production of useful products such as acetic acid (Shanableh 2000), biofuel from microalgae (Garcia-Alba et al. 2013) and synthesis of methyl methacrylate (Giudici and Maugans 2000). The main reactions are similar to incineration, and any substance that can be incinerated can be oxidized in water via WAO. The WAO process is therefore perfect for treating waste liquors, slurries and sludge where the organic matter is very high in concentration compared to water. Another benefit of the WAO process is that nitrous oxide, sulfur dioxide, hydrochloric acid, dioxins, furans, and fly ash are not produced. This process is capable of up 99 % conversion of toxic organics to harmless end products. For compounds which are not completely oxidized, intermediate compounds representing up to a quarter of the original mass of the organic matter are produced, such as small carboxylic acids. Typical conditions for WAO are:

- Temperatures- between 150 and 320 °C,
- Pressure – 20–150 bar
- Residence time – 15–120 min.

The type of application is usually determined by the range of temperatures used. Low temperature oxidation −100–200 °C is used for thermal conditioning of municipal and paper industry sludge, whereas medium temperature −200–260 °C oxidation is typically used for treatment of ethylene spent-caustics and some other industrial wastes, and also for regeneration of powdered activated carbon used in wastewater treatment. Higher temperatures −260–320 °C are used for sludge destruction and treatment of industrial wastewaters including organic industrial wastes such as pharmaceutical wastes and solvents. At the higher end of this temperature range, complete destruction of municipal, pulp and paper and other organic sludge is expected (Giudici and Maugans 2000). This high temperature range is within the sub-critical region for water where the solubility of salts is reduced. Precipitated salts may be the cause of the corrosion that was a problem for early Zimpro sludge treatment operations. The process of WAO must be conducted in the aqueous phase so high pressures are required to maintain water as a liquid. Pressurization also increases the concentration of dissolved oxygen and thus increases the oxidation rate (Debellefontaine and Foussard 2000).

This process is similar in principle to SCWO with the exception that the reaction mixture is kept below the critical point of water. While SCWO can achieve complete oxidation of the organic fraction of the sludge solids, wet air oxidation can achieve effective hydrolysis (>95 % as COD) of the sludge organic compounds but incomplete oxidation (<95 % as COD) (Shanableh 2000). Process efficiency depends upon operating parameters such as temperature, pressure, air supply and feed solids concentration. The extent and the rate of oxidation can be increased by elevating the reaction temperature within the range 120–370 °C. Operating pressures between 1 and 27 MPa can be used depending upon the degree of oxidation desired. As with incineration, an external oxygen supply is needed. Since thermal efficiency and process economics strongly depend on air input, it is thus very much important that the optimum air requirement is determined. As an example, activated sludge with a heat value of 15,212 kJ/kg (6,540 BTU/lb) typically requires 5.14 kg air/kg of sludge for wet air oxidation (US EPA 1979). Although wet oxidation does not require predewatering (as low as 1 % solids sludge can be fed to the process), the process operating costs can be significantly reduced by increasing sludge consistency. In one study wet air oxidation costs decreased from $38 to $23 tonne–1 by increasing sludge solids from 3 to 6 %. High sludge solids keep the oxidation process self-sustaining.

The wet air oxidation process has been commercialized as the Zimpro Process (Mahmood and Elliott 2006). The major installations of the Zimpro process were:

- The Chicago Sanitary District's installation at its West Southwest Wastewater Treatment Plant (US EPA 1979)
- The Springfield Water and Sewer Commission's regional activated sludge wastewater treatment facility located on Bondi Island (Borgatti et al. 2000).

The Chicago installation was operated for 10 years, after which, the Zimpro process was withdrawn in favour of a landfill disposal system. The Springfield Water and Sewer Commission's facility obtained a 50 % destruction of volatile sludge solids using the Zimpro process since its installation in 1970. Later energy intensive Zimpro process was replaced with an extended aeration operation of the upgraded ASP, which, since then, has reduced secondary sludge production by 30 % (Borgatti et al. 2000).

Wet air oxidation has several advantages (Collyer et al. 1997):

- Air pollution-free operation
- Far lower ash production in comparison to incineration
- No need of sludge dewatering and thus savings in sludge conditioning costs

A major weakness of the wet air oxidation is the fact that it produces high-strength liquors, which need to be recycled to the treatment plant (Shanableh 2000) requiring increased aeration capacity. BOD of the liquor can be as high as 40–50 % of that of the raw sludge, which translates to a 30–50 % increase in BOD loading to the treatment system. Both surface and subsurface reactor designs have been recommended to achieve super and subcritical water oxidation. Surface reactors are high-pressure vessels. These are capable of sustaining high pressures applied by mechanical devices such as pumps. A subsurface reactor is constructed under ground; deep enough that the hydrostatic head achieves the required pressure. If the entire pressure required for the SCWO is to be developed by the hydrostatic head, a reactor depth of approximately 3,650 m would be required (US EPA 1994). Based on similar principles, a wet oxidation technology which is also known as deep-hole technology has been developed in Netherlands for sludge oxidation. This consists of a set of vertical pipes, which are drilled vertically about 1,500 m from the surface of the Earth. Pure oxygen is supplied to start-up the process and afterwards, heat is generated from the exothermic sludge oxidation. The liquid is biologically treated while the ash is dewatered for disposal. The technology is not suitable for use in some geological conditions and still has a long way to go to prove its economic viability (De Bekker and Van den Berg 1988; Spinosa et al. 1994).

Recently, work has been done on developing wet oxidation under milder reaction conditions and lower pressure (Abe et al. 2011, 2013). Wet air oxidation treatment at 150 °C temperature, 10 bar pressure and 2 h reaction was able to give a 62 % volatile suspended solids removal efficiency. This lower pressure WAO process was considered as a sludge pre-treatment process to improve the sludge characteristics for anaerobic digestion. Studies suggested that an excessive concentration of oxygen used in the reaction led to production of recalcitrant soluble organics and toxic compounds and can reduce gas production in anaerobic digestion (Abe et al. 2013). Typically, catalysts will lower reaction temperatures and pressures to be used to achieve the same results as those achieved without catalysts in WAO processes. Refractory compounds such as acetic acid and ammonia also become more susceptible to oxidation (Luck 1999).

5.4 Super Critical Water Oxidation

Supercritical water oxidation (SCWO) is basically an evolution of the WAO process where the operating temperature is increased beyond the critical temperature of water (Blaney et al. 1995). However, because supercritical water behaves very differently to sub-critical water, many of the reactions and mechanisms in SCWO would be different from WAO. The SCWO process has been used for the treatment of various wastes (Brunner 2009) including sludge. SCWO is showing promise and is being continually developed for application in sludge treatment. Supercritical Water Oxidation, also referred to as hydrothermal oxidation, is a process that oxidizes organic solutes in an aqueous medium using oxygen/air or hydrogen peroxide as oxidants, at temperatures and pressures above the critical point of water, i.e., 374 °C and 22 MPa, respectively (Bermejo et al. 2006).

The SCWO process has been under development since the early 1980s when the well known process of wet oxidation was developed at MIT (Modell 1982). SCW is a superior reaction medium with a high diffusivity, a low viscosity, and relatively high-density therefore rapid oxidation reactions are expected. Moreover, the low temperature of the SCWO process in comparison to conventional combustion can lead to a greatly reduced NOx and SO_2 formation. In addition, water is not only the reaction medium but it participates directly in the reaction through the formation of free radicals (Griffith and Raymond 2002). Since water is utilized in the reaction there is no requirement to dewater the sludge before processing. Sludge can be processed at 10 % solids by weight or even less (Mahmood and Elliott 2006).

Figure 5.2 presents a schematic of a SCWO process. Pressurized sludge and pressurized oxygen are fed into the preheater reactor at 25°°C. Sludge is pressurized to 25.5 MPa. In the preheater the mixture of sludge and oxygen are heated up to approximately 300–400 °C, to achieve the supercritical state of water. Water at its

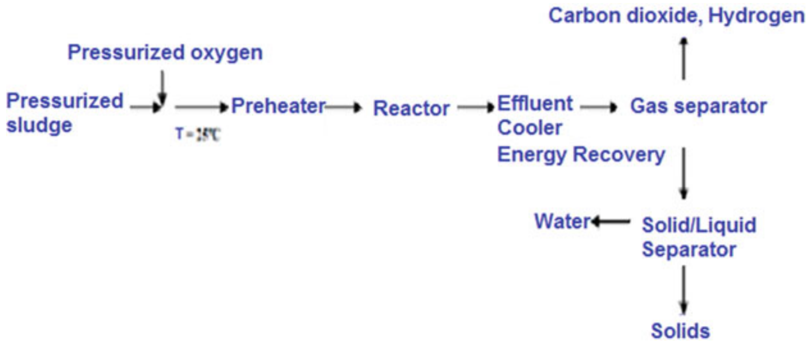


Fig. 5.2 Schematic of a supercritical water oxidation system (Based on Bambang and Jae-Duck 2007, Xu and Lancestor 2009, Mahmood and Elliott 2006)

supercritical state can dissolve organics and hydrolyze even polymers and hence prevent the formation of char (Perry and Green 1999; Fang and Koziński 2000; Mahmood and Elliott 2006). The reaction mixture enters the main reactor where the remaining portion of the organics is oxidized in short hydraulic residence time of 5–10 min at the maximum process temperature of around 600 °C (Mahmood and Elliott 2006). After reaction, the effluent is cooled and energy is recovered. According to Svanstrom et al. (2004), about half of the heating value of the sludge can be recovered in the studied process. The solid and liquid products are separated and the wet inorganic solids can be sent to a landfill or spread on dedicated land while the water can be redirected to the wastewater treatment plant. The efficacy of SCWO has been shown at the laboratory and pilot scale with a wide broad range of feedsctocks, such as pig manure, a variety of biomass slurries including pulp mill sludge and sewage sludge (Rulkenes et al. 1989; Modell 1982, 1990; General Atomics 1997). It has been demonstrated that complete oxidation of almost any organic material, including hazardous wastes such as hexachlorobenzene, could be obtained by using the SCWO process.

At Harlingen, Texas, USA, a supercritical water oxidation sludge processing plant has been installed to process up to 9.8 dry tonnes per day of municipal sludge (Griffith and Raymond 2002). An environmental assessment was conducted on the Harlingen plant and found huge environmental gains from recovery of heat and thus reducing natural gas consumption for heat generation (Svanström et al. 2004). Hydrothermal oxidation, in particular SCWO, is currently being considered by several research and waste management organizations as an alternative treatment option (Stark et al. 2006). Figure 5.3 shows the flowchart of the SCWO pilot plant (Gidner et al. 2001).

This process can very efficiently oxidize organics at moderate temperatures (400–650 °C) and high pressure (25.5 MPa). Laboratory scale studies have demonstrated total organic carbon and AOX reductions of 99–99.9 % and dioxin reductions of 95–99.9 % (Modell 1982). Optimum SCWO results were achieved at 650 °C (Murakami 1998). Complete removal of organic carbon was achieved at this

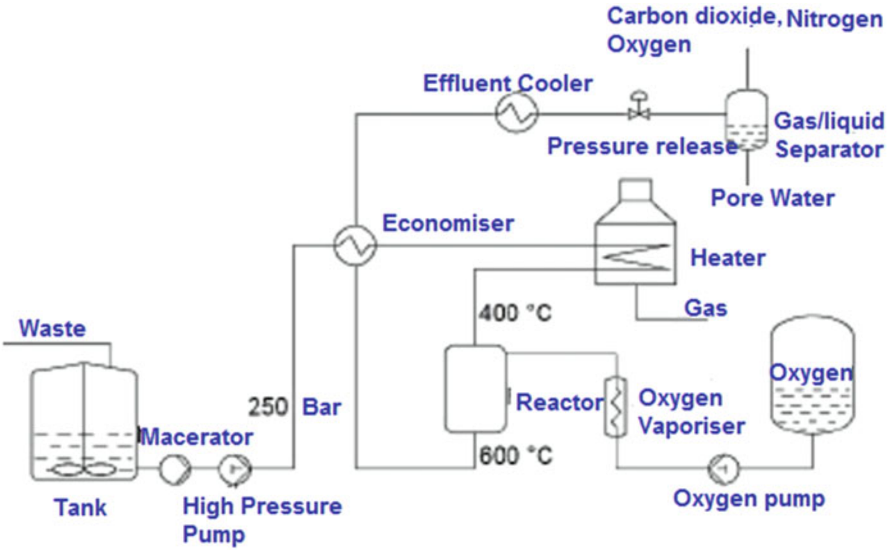


Fig. 5.3 Flowchart of the supercritical water oxidation pilot plant (Based on Deviatkin 2013, Gidner et al. 2001)

temperature in 10 s and the exhaust gas did not contain any NOx and SOx. One advantage of this process is that it does not require a high degree of dewatering prior to oxidation. Sludge can be processed at 10 % solids by weight or even less. Modell (1982) have reported that SCWO process is cheaper than dewatering plus incineration for treating pulp mill sludges because of regenerative heat exchange to preheat feed and cool effluents. Also, this process was found to compete effectively with dewatering plus landfilling costs where tipping fees exceed U.S. $45 m–3 (Modell 1982).

Cooper et al. (1997) treated primary clarifier sludge at 30 % solids mixed with bleach plant effluent (30 % Do:70 % EOP) at 7 % solids and decant pond sludge by SCWO. The sludges were wet ground before oxidation in order to reduce particle size below 0.15 mm. It was found that the two sludges were oxidized at 81.4 % and 97.7 % organic carbon destruction efficiencies and 99.47 % and 99.93 % AOX removal efficiencies, respectively. The liquid effluents from the two sludges were colourless and neutral in pH but contained 460 and 420 mg/L dissolved organic carbon (DOC), respectively.

Dahlin (2002) and Gidner and Stenmark (2002) reported how SCWO can be effective at treating deinking sludge from a pulp and paper facility. The process oxidizes organics and it also allows the recovery of a paper filler. The trade name of the process is Aqua Critoxs (Minett and Fenwick 2001).

Izumizaki et al. (2005) reported the production of hydrogen from paper sludge in supercritical water in the presence of ruthenium (IV) dioxide, RuO2. The reaction conditions were 100 mg paper sludge, 20 mg catalyst, temperature of 450 °C for 2 h. The major components of gases produced were hydrogen, methane and carbon dioxide in molar ratios of 27 %, 27 % and 45 %, respectively. Canadian researchers have started an extensive study on catalytic gasification of pulp/paper secondary sludge in SCW for hydrogen production (Xu and Lancestor 2009).

WAO process is able to produce volatile fatty acids, mainly acetic acid, which can be recovered for use (Hii et al. 2013). The production of acetic acid using WAO at sub-critical conditions was studied by Shanableh (2000) for use as the organic reactants necessary in denitrification processes in WWTPs. The WAO efficiently hydrolyses sludge solids but achieves incomplete oxidation of the organic components. This produced COD-rich liquors containing 10 % wt/wt acetate. This accounted for up to 80 % of soluble COD. Chung et al. (2009) found that the formation of organic acids increased with reaction temperature. More organic acids were produced as intermediates when the reaction temperature was increased. Acetic acid production increased by four times as temperature was increased from 180 to 240 °C, at 40 min reaction time. Strong et al. (2011) were able to obtain a slightly higher production of acetic acid (15 % wt/wt) using WAO process at 220 °C.

The SCWO process has been studied at a pilot scale in Germany on several sludges and on one deinking sludge in Sweden. The German tests on primary sludge gave very high removals of chlorinated organics (>99 %), but comparatively low overall organics removal (98 %). The Swedish work is being carried out by Chematur using its pure oxygen-based Aqua Critox process, which is being jointly marketed with BOC. Pilot trials carried out at Stora Enso's Hylte newsprint mill in Sweden have produced about 5 tonnes of recovered filler, which has been used in trial newsprint runs with no adverse effects on paper quality (Schmieder and Abeln 1999). The advantages of both WAO and SCWO over conventional incineration is that the emissions to air are very low because any sulphur, nitrogen and chlorine compounds remain in the liquid phase.

The comparatively lenient operating condition of the subcritical hydrothermal treatment (HTT) would be more practical, which has been investigated by several researchers in recent past (Namioka et al. 2009; Prawisudha et al. 2012; Muthuraman et al. 2010; Nakhshiniev et al. 2012; Sakaguchi et al. 2008; Indrawan et al. 2011). According to the previous successful studies and results, the HTT could be successfully implemented to the paper sludge. Areeprasert et al. (2014) investigated the solid fuel production from paper sludge employing HTT in a lab-scale facility for implementation of the pilot-scale plant. The paper sludge was subjected to the HTT under subcritical hydrothermal conditions. In the lab-scale experiment, the temperature conditions were 180 °C, 200 °C, 220 °C, and 240 °C, respectively, while it was 197 °C in the pilot plant as the optimum condition. The holding time was 30 min in both cases. The hydrothermally produced solid fuel was evaluated for the fuel property, the water removal performance, and the mass distribution. Furthermore, the energy balance of the process was studied. The higher heating value of the HT pretreated paper sludge was slightly improved. In addition, the produced solid fuel had comparable H/C and O/C atomic ratios with that of coal, indicating the presence of carbonization during the HTT process. Using the mechanical dewatering, only 4.1 % of moisture in the raw paper sludge can be removed while the solid fuel production from paper sludge by HTT at 200 °C

Fig. 5.4 Deinking sludge before and after SCWO (Reproduced with permission Kay 2003)

showed 19.5 % moisture reduction. According to the energy balance of the pilot plant, the recovered energy was significantly higher than the energy input, showing the feasibility of employing the HTT to produce alternative solid fuel from paper sludge.

Figure 5.4 shows Deinking sludge before and after SCWO treatment (Kay 2003).

5.5 Steam Reforming

Steam Reforming technology is used for sludge treatment. It is still considered as an emerging technology for paper sludges. Steam reforming is a novel combustion technology, which carries out in a steam reforming reaction system (Durai- Swamy et al. 1990, 1991; Aghamohammadi and Durai-Swamy 1995; Demirbas 2007). Steam reforming is based on an innovative pulse combustion technology carried out in a steam reforming reactor system. Pulse combustion is a phenomenon of combustion induced oscillations which are incorporated by design to achieve a high heat release and more complete combustion. The technology not only offers enhanced heat transfer rate but also low NOx emissions. In addition, operating the steam reformer at a lower temperature (500–600 °C) minimizes the vaporization of toxic metals which remain in the char (Durai-Swamy et al. 1990, 1991; Aghamohammadi and Durai-Swamy 1995; Demirbas 2007). Manufacturing and Technology Conversion International (MTCI) has developed a unique process that reduces the volume of the solid wastes, destroys the chlorinated organics such as dioxins, and produces clean fuel gas for use in the mill as a replacement for natural gas. The test results confirmed the ability of the MTCI indirect gasifier to handle a wide variety of biomass feedstocks, including those with high moisture content, low ash fusion temperature, and high plastic materials content. Also, product gas quality was shown to be quite insensitive to feedstock moisture level. The gasifier does not require any special feedstock preparation such as pelletization. The gasifier produces a medium-Btu gas without the consumption of oxygen. The reactor is easily scaled, since the pulse combustor tube bundles are constructed in modules. The gasifier produces a gas with a hydrogen-to-carbon-oxide ratio significantly higher than oxygen-blown systems, thus making it particularly attractive for methanol production. The gasifier integrates well with both methanol and high purity hydrogen plants. The results of these tests have provided a significant database for preparing designs at the 50 tons/day level. MTCI plans to field demonstrate the gasifier at a commercial paper mill in the near future.

5.6 Gasification

Application of gasification technology to pulp and paper is considered to be in its early stages of development and there are very few commercial installations (US EPA 2012). It is a thermal process during which a combustible material is converted into an inflammable gas and an inert residue using air or oxygen. So far, gasification of coal is the most abundantly applied form of gasification. According to US EPA (2012), the last 10–15 years have seen the rebirth of gasification technology (Fig. 5.5). There are several reasons for this. The most important reason is the remarkable increase in energy costs. Oil prices were between $20 and 30 per barrel before 2003. Prices have mostly been in the range of $55–120 per barrel since 2005. With natural gas, the commercial price from 1983 to 2003 was mostly between

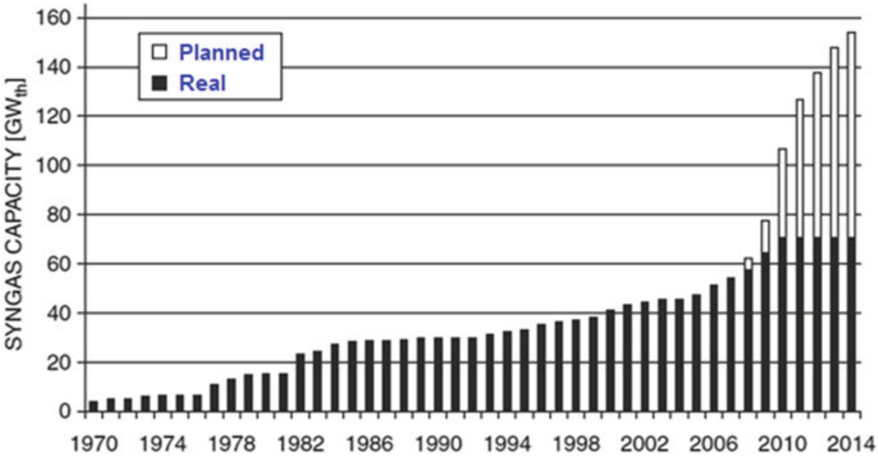


Fig. 5.5 Wordwide gasification capacity (Based on US EPA 2012)

$5 and 6 per MMBtu, rising slightly toward the end of the period. It remained consistently over $10 per MMBtu, and reached at $15 at the end of 2005 in United States between 2005 and 2009 (Higman and Burgt 2008). According to Roos (2008), more than 100 biomass gasifier projects are operating or ordered in different parts of the world. The gasification of woody biomass is being looked at due to the

- Availability of timber in large quantities in the United States and other areas
- Renewable energy standards and goals
- Greenhouse gas emission regulations and reduction goals

Thermal and electrical energy, and also liquid fuels can be produced by gasification of woody biomass. As these processes become more widespread, proven, and well accepted, similar systems processing more complex feedstocks are expected to be demonstrated and commercialized, including those for sludges and municipal solid waste.

The basic principle of gasification is to convert a carbon based material into hydrogen and carbon monooxide with the addition of heat and a combination of steam, oxygen and/or nitrogen in a reaction vessel (US EPA 2012). Other than hydrogen and carbon monooxide, the remainder of the syngas includes nitrogen, traces of methane and other hydrocarbons, tar, particulates, and carbon dioxide. Once produced, the syngas can be cleaned through the use of a variety of cleanup devices. These include:

- Ash-capturing cyclones,
- Solvent based tar scrubbers
- Water, acid or caustic scrubbers for capturing nitrogen, chlorine, sulfur and various heavy metals

After cleaning, the syngas can be converted to a liquid fuel using a catalytic Fischer-Tropsch (FT) process, fed into an internal combustion engine-generator for electricity production, combusted for heat recovery, used in fuel cell applications, or used for the production of a wide variety of chemicals (US EPA 2012). Theoretically, any form of biomass may undergo gasification. Limitations on the efficiency of the gasifier's operation include several factors listed below (Sadaka 2008):

- High moisture content of the feed
- Ash fusion temperatures
- Design of the feeding system
- The mixing and separation of the feedstock

Feedstock is first prepared by drying to the appropriate moisture content, typically between 10 and 20 %. After drying, the feedstock is then transferred to a feeding system, which can vary in design based upon the pressure in the gasifier and the physical properties of the feedstock. After entering the gasifier, the feedstock is then converted into syngas of varying compositions, which depends on the gasifier type and feedstock composition. A cyclone installed downstream of the gasifier will capture additional ash/particulate matter that is not captured in the gasifier. Heat can be recovered in the form of steam with the use of a

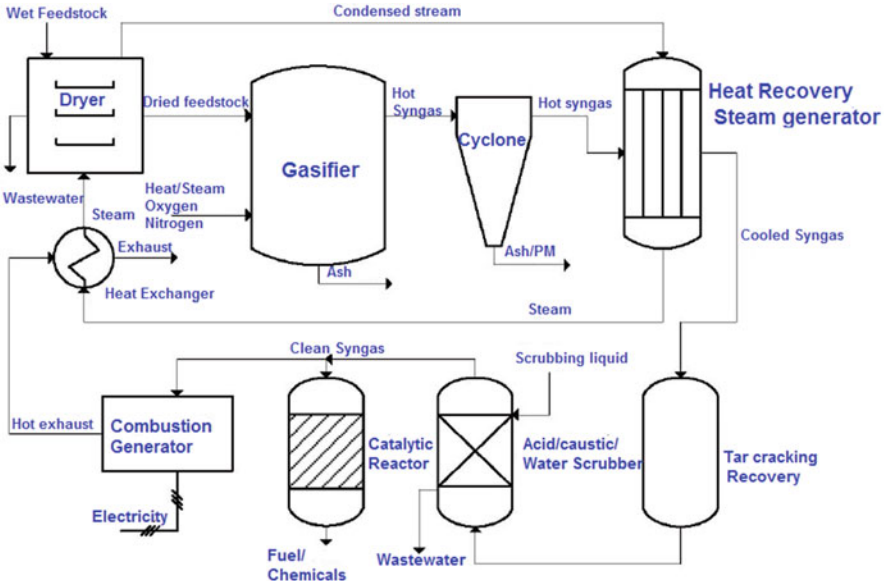


Fig. 5.6 Gasification system (Based on US EPA 2012)

heat recovery steam generator and fed into the dryer to supplement the drying system. The cool raw syngas is then treated through a liquid and/or dry cleaning system. Finally, the cleaned syngas is then fed into the conversion system to create electricity, heat, fuels, and/or chemicals. If using a combustion generator, additional thermal energy can be removed from the exhaust to further supplement the drying system. In Fig. 5.6, a generic system is presented. In addition to the value of the end products, the availability of the feedstock, pretreatment requirements, gasification system efficiency, syngas conversion process and site specific energy costs all have a significant effect on whether or not a system will be commercially successful.

Several types of gasifier designs are presently being used for commercial applications in the production of fuels and chemicals. Gasifiers applicable to sludges include fixed bed, fluidized bed and those grouped as plasma/other. A summary of some of the main aspects of the main gasifier types are presented in Table 5.2.

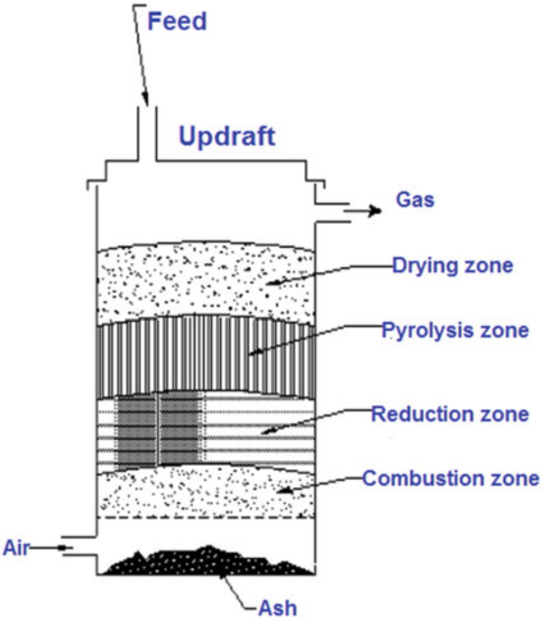
The oldest form of gasification is the fixed-bed process. In the Fixed-bed updraft gasifiers there is a bed in which the feedstock moves slowly downward under gravity as it is gasified by a gasification medium which is in a counter-current flow to the feedstock. In such a arrangement, the hot syngas from the gasification zone is used to preheat and pyrolyze the downward flowing feedstock. In case of a fixed-bed downdraft gasifier, the flow of the feedstock is concurrently with the gasification medium. Figures 5.7 and 5.8 shows the updraft and downdraft process (US EPA 2012). In a fluidized bed gasifier, the gasification medium and feedstock must pass through a bed of inert particles (e.g., alumina oxide). There are two types of fluidized bed gasifiers:

Table 5.2 Key aspects of the main gasifier types applicable to sludges

Bubbling fluidized bed gasifier	Medium scale; higher throughput; reduced char; ash does not melt; simpler than circulating bed
Circulating fluidized bed gasifier	Medium to large scale; higher throughput; reduced char; ash does not melt; excellent fuel flexibility; smaller size than bubbling fluidized bed
Downdraft fixed bed gasifier	Small scale; easy to control; produces biochar at low temperatures; low throughput; higher maintenance costs
Updraft fixed bed gasifier	Small and medium scale; easy to control; can handle high moisture content;·low throughput
Supercritical water	Short reaction time; high energy conversion efficiency by avoiding the process of drying step; selectivity of syngas with temperature control and catalysts
Plasma	Large scale; easy to control; process is costly; high temperature (5,000–7,000 °F)
Liquid metal	High syngas quality

Based on US EPA (2012)

Fig. 5.7 Fixed bed updraft gasifier (Based on US EPA 2012)



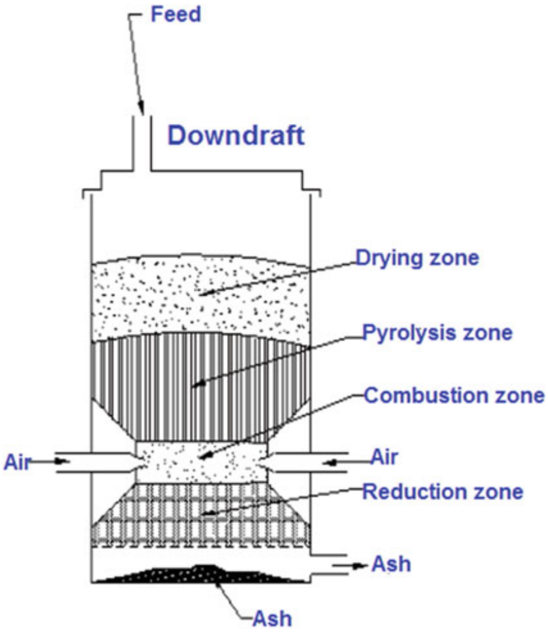
– Bubbling fluidized bed gasifiers:

These are typically appropriate for medium size projects of 25 MWth or less,

– Circulating fluidized bed gasifiers:

These gasifiers can range from a few MWth up to very large units.

Fig. 5.8 Fixed bed downdraft gasifier (Based on US EPA 2012)



There is extremely good mixing between feedstock and oxidant in Fluidized bed gasifiers. These gasifiers promote heat transfer and mass transfer. This ensures an even distribution of material in the bed, but a certain amount of partially reacted fuel is inevitable and will be removed with the ash. This places a limitation on the carbon conversion of fluid-bed processes. The operation of fluidized bed gasifiers is generally restricted to temperatures below the softening point of the ash, since ash slagging/agglomeration will disturb the fluidization of the bed (Higman and Burgt 2008). Figures 5.9 and 5.10 shows the bubbling and circulating bed processes (US EPA 2012).

Plasma gasification is a fairly new process with respect to traditional gasification (US EPA 2012). The primary heat source in a plasma gasifier is the plasma torch, where gas is passed through an electric arc and dissociated into ions and electrons creating extremely high temperatures. The high temperatures facilitate very large carbon conversion percentages and also good control of the hazardous materials captured in the slag; however, plasma gasifiers are relatively costly and have relatively higher parasitic energy consumption when compared to traditional gasifiers (For more details see Sect. 5.6.1).

Some companies are investigating liquid metal gasification at pilot scale. Feedstock is introduced into a crucible filled with molten metal which is usually iron, at around 1,300 °C. Water in the feedstock is converted into hydrogen and oxygen. The iron is then oxidized to iron oxide and then reduced back to iron after the oxygen reacts with carbon in the feedstock to make carbon monooxide gas. The hydrogen and carbon monooxide gas are the main two components in the syngas. In order to

Fig. 5.9 Fluidized bed gasifiers – bubbling bed (Based on US EPA 2012)

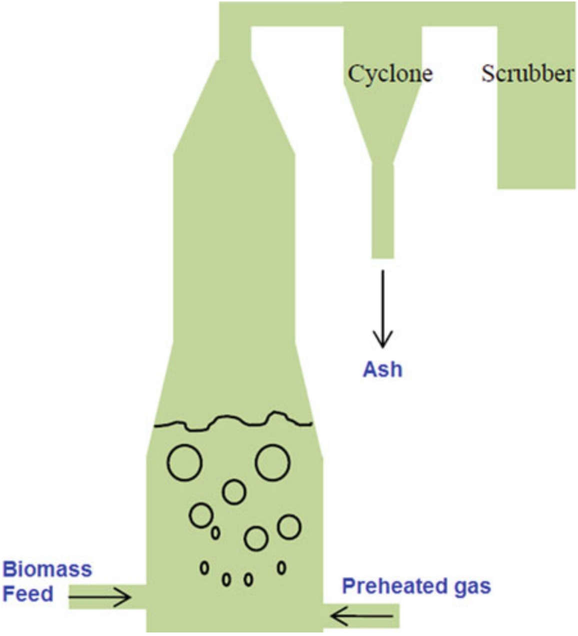
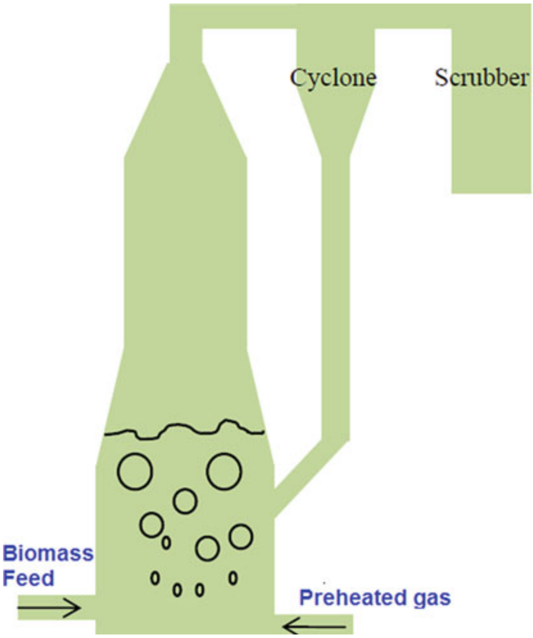


Fig. 5.10 Fluidized bed gasifiers – circulating bed (Based on US EPA 2012)



favorably shift the equilibrium, oxygen gas can be introduced. The iron also helps to capture unwanted waste like chlorine and sulfur into a glass like material which is called slag.

Supercritical Water Gasification is a process which uses super critical water (pressure more than 320 psi, temperature more than 600 °F) to convert organics into a hydrogen rich syngas. This process requires feedstocks with moisture contents ranging from 70 to 95 %. The reforming of biomass and biological residues in supercritical water is a rather novel process. Significant research will be required for commercialization of this technology (Biomass Technology Group 2010) (For more details see Sect. 5.6.2).

Pyrolysis can also be considered as a gasification process, but it is conducted in the absence of oxygen. Both processes may also be performed together. Gasification can be applied to the solid residue of the pyrolysis. It is a new method when applied to the sludge, and therefore not very well known. The input to the process can either be digested or undigested mechanically dewatered sludge (European Commission 2001; CANMET 2005).

During gasification, sludge undergoes a physical and chemical change, similar to other biomass feedstocks. Because of the high moisture content, it is important for the sludge feed to be dewatered in some way before entering the reactor. This is true for most reactors, although some technologies that operate at much higher temperatures, such as plasma gasification, have the ability to handle sludge without pretreatment. The gasification process itself will not be affected by the high moisture content, but because of the high energy demand required by the system to vaporize the moisture, the capacity and economics are affected (Sadaka 2008).

The largest obstacle related to sludge gasification is reducing the water content to a level suitable for gasification. Mechanical processes are preferred to thermal processes from an energy standpoint, but secondary sludge can only be mechanically dewatered to about 40 % solids (Cheremisinoff 2002). The difficulty in removing water from secondary sludge is due to the water trapped inside the cell walls of the organisms used to consume the biological oxygen demand of the wastewater that remains after the primary settling. The cell walls must be broken to remove this moisture.

Paper mill sludges can be reduced to 50 % moisture by using a belt press followed by a screw press (Kandaswamy et al. 1991). To finish the preparation process, thermal energy is required to remove the remaining moisture. Biosolids are the nutrient-rich organic materials resulting from the treatment of sewage sludge (the name for the solid, semisolid or liquid untreated residue generated during the treatment of domestic sewage in a treatment facility). Sewage sludge becomes biosolids when treated and processed to achieve the pathogen and/or pollutant limits set forth by the EPA's Part 503 Biosolids Rule. The limits can vary based on the biosolids use or disposal and classification (US EPA 1994).

In a gasification situation, composting and lime stabilization techniques would be eliminated. Aerobic digestion would also be eliminated. Anaerobic digestion, although compatible with gasification, is not suitable as a pre-treatment step because much of the chemical energy of the biosolids is removed and so not available for recovery in the syngas. After the treated sludge is dewatered then dried to a low enough water content (80–90 % solids), inorder to be properly gasified, the dried sludge enters the gasifier chamber, and undergoes the first step of gasification in a typical fixed bed downdraft gasifier (Sadaka 2008; US EPA 2012).

In the drying zone, the sludge moves down into the gasifier and moisture is evaporated using the heat generated in the zones below. The rate of drying depends on the:

- Surface area of the fuel
- Recirculation velocity
- Relative humidity of these gases
- Temperature differences between the feed and hot gases
- Internal diffusivity of moisture within the fuel

Sludge with less than 15 % moisture, loses all moisture in this zone (Sadaka 2008). In the pyrolysis zone, the irreversible thermal degradation of dried sludge descending from the drying zone takes place using the thermal energy generated by the partial oxidation of the pyrolysis products. The volatiles are released from the sludge at about 250 °C, and 60–70 % of sludge is converted to a complex liquid fraction containing water, tars, oils, a gaseous phase and a variety of other hydrocarbons, and un-reacted char and ash. Gaseous phase include carbon dioxide, carbon monooxide, hydrogen. It is expected that pyrolysis of sludge in a reactor typically occurs at temperatures between 350 and 500 °C (Sadaka 2008). The throat zone is often referred to as the oxidation zone. In this zone, the volatile products produced from the pyrolysis process are partially oxidized in highly exothermic reactions, which result in a rapid rise in temperature up to 1,100 °C. The heat generated is used to drive the drying and pyrolysis of sludge and the gasification reactions. The oxidation reactions of the volatiles are very rapid and the oxygen is consumed before diffusing to the surface of the char. Therefore, no combustion of the solid char can take place. Oxidation of the condensable organic fraction to form lower molecular weight products is important in reducing the amount of tar produced. During sludge gasification, the oxidation zone temperatures are between 1,000 and 1,100 °C. The products, including carbon dioxide, carbon monooxide, hydrogen, water, high chain hydrocarbon gases, residual tars and char, then pass on into the gasification zone (Sadaka 2008). The reduction zone is often referred to as the gasification zone. In this zone, the char is converted into gas by reaction with the hot gases from the upper zones. The gases are reduced to form a larger proportion of carbon monoxide and hydrogen. Temperatures of the gases entering this zone are about 1,000–1,100 °C and exit around 700 °C (Sadaka 2008).

The energy content of typical pulp and paper mill sludge is around 3,600 Btu/lb, dry. If sludge at 10 % moisture is fed into a fixed bed air blown gasifier, the syngas energy content would be approximately 130 Btu/scf. If syngas coming out of a 5 ton/day gasifier is sent to an electrical generator with a 40 % electrical efficiency, 108 kW of gross electricity could be produced. Taking into account a parasitic load from the gasifier, dryer, and cleaning system of approximately 75 kW, a positive net output is possible, but it is very difficult to obtain.

The major technical challenge for sludge gasification is associated with ash slagging and the formation and removal of tar which is a high molecular-weight hydrocarbons rich in benzene, toluene and xylene. The presence of tar in the producer gas is not desirable because of the following reasons (Rezaiyan and Cheremisinoff 2005):

- It is an indicator of low gasification efficiency
- It increases the difficulty of syngas cleanup by fouling and plugging the pipes and tubes of some equipment

Therefore, it is necessary to develop technical approaches to remove tar. Tar removal can be achieved either by primary method taking place insider the gasifier or by secondary treatments outside the gasifier. As a primary method, choosing the proper configuration of a gasifier can reduce tar formation. Brandt and Larsen (2000) used a novel two-stage gasifier composed of a pyrolysis unit and a gasification unit with a charcoal bed and produced a significantly low tar formation. Another research group also observed a reduction in tar formation when the producer gas went through a second-stage bed packed with char in a downdraft fixed-bed gasifier (Nunes et al. 2007). Addition of catalysts, such as char, alkali/alkaline earth metal-based catalysts (sodium, potassium and calcium) and transitional metal-based catalyst (nickel and iron), proved to be an effective means to reduce tar formation by converting tar into combustible gases through steam reforming, carbon dioxide reforming, thermal cracking, hydro-reforming/hydro-cracking, and water-gas reactions (Kimura et al. 2006). The secondary method for tar removal, hot gas cleanup has attracted significant attention in recent years due to the development of integrated gasification combined cycle (IGCC) and integrated gasification fuel cell (IGFC) technologies. Hot gas cleanup, which is catalytic destruction of tarry products and ammonia (a contaminant species in the producer gas) at a high temperature is needed to further increase the overall power generation efficiency of IGCC and IGFC. The most common catalysts for the decomposition of tar and ammonia are dolomite (a calcium magnesium ore, $CaMg(CO_3)_2$) and nickel, molybdenum, or ruthenium based catalysts (Dayton 2002), and also an inexpensive iron catalysts such as chars from low rank coals with inherent iron and calcium cations and limonite iron ore (Ohtsuka et al. 2004; Xu et al. 2005).

Durai-Swamy et al. (1990) used a pulse-enhanced, indirectly heated fluidized bed gasifier system to treat wood chips, recycle paper mill sludge and Kraft mill sludge. Carbon conversions to dry gas were found to be 93 %, 87 % and 80 % for wood chips, recycled mill fibre waste and Kraft mill sludge, respectively. The respective char productions from carbon were 4 %, 8.5 % and 19 %. A small fraction of sludge organics was converted to oil. Wood chips, recycled mill fibre waste and Kraft mill sludge were processed wet as received at moisture contents of 25 %, 50 % and 62 %, and ash content of 0.2 %, 3 % and 19 %, respectively. Dioxin emission from sludge incineration has been a concern. The technology developed by Durai-Swamy et al. (1990) performs gasification in a bed of calcium-based material, which is reactive towards chlorine containing dioxin precursors. The technology has thus been claimed to suppress the dioxin formation potential (Durai-Swamy et al. 1990). Krause and Levert (2004) are marketing the deep bed bark/sludge combustion process which incorporates gasification to maximize power generation from the combustion of wet fuels from pulp and paper mills.

Ouadi et al. (2013) explored the gasification of blends of pre-conditioned rejects and de-inking sludge pellets with mixed wood chips in an Imbert type fixed bed downdraft gasifier with a maximum feeding capacity of 10 kg/h. The producer gases evolved would generate combined heat and power (CHP) in an internal combustion engine. The results show that as much as 80 wt% of a brown paper mill's rejects consisting of 20 wt% mixed plastics and 80 wt% paper fibres could be successfully gasified in a blend with 20 wt% mixed wood chips. The producer gas composition was 16.24 % hydrogen, 23.34 % carbon monooxide, 12.71 % carbon dioxide 5.21 % methane and 42.49 % nitrogen (v/v %) with a higher heating value of 7.3 MJ/Nm^3. After the removal of tar and water condensate the producer gas was of sufficient calorific value and flow rate to power a 10 kWe gas engine. Some blends using rejects from other mill types were not successful, and the limiting factor was usually the agglomeration of plastics present within the fuel.

There is difference in the chemistry of pyrolysis and gasification technologies as operating temperature and control of oxidation by air are different in these two processes. Pyrolysis requires an extensive residence time to achieve an optimum quality of char. Gasification technologies optimize the production of produced gasses and pyrolysis technologies optimize the production of char, as well as heavy and light oils (CANMET 2005).

Unlike the incineration process, gasification is much more dependent on fuel characteristics. The major criterion for successful gasification is moisture content of the fuel. In general terms, it is possible to gasify sludge with 55 weight-% moisture, while it is recommended to dewater and dry sludge up to only 10 weight-% moisture content to provide optimum efficiency of the process. Sludge moisture affects the reactor efficiency to negative side, and also decreases the quality of syngas (CANMET 2005). With the development of waste-to-energy technologies market, the range of equipment available expands. Different configurations of already existing facilities and new emerging technologies are ready to be applied. All gasification technologies can be grouped into the next categories: circulating fluidized bed boiler, fluidized bed boiler, fixed bed, updraft and downdraft, static and rotating beds. In recent times, plasma technology and supercritical water gasification technologies are becoming popular (Xu and Lancaster 2012).

Gasification process is becoming more and more popular because of several benefits gained: firstly, volume of flue gases and concentration of pollutants therein is lower than that of incineration process; secondly, overall energy utilization efficiency of the process is higher. Operating temperature of the process ranges between 900 and 1,400 °C (Xu and Lncaster 2012). Compared to conventional methods, gasification offers a potentially viable option for sludge disposal and is capable of providing a clean and manageable process with the possibility of net energy gains. Unlike incineration, there is potential for sludge gasification to deliver negative GHG emissions. The magnitude of GHG reductions is highly site, technology and/or feedstock specific. Because of this, a general statement cannot be made that identifies gasification as having a lower carbon footprint than other management practices.

Several companies claim to be able to gasify sludge, but supporting independent data on their processes is not available. In addition, many different system uses and designs are available, even among the handful of early commercial systems. As a result, a complete technical and economic analysis will only be feasible for this technology and industry when implemented more broadly through a case by case basis analysis. More specifically, when a pretreatment process, gasifier, clean up system and energy recovery process have been integrated and commissioned, the system can be thoroughly evaluated through collected data. Based on the quantity of research data pertaining to sludge gasification, it is evident that there is significant interest all over the world in developing this technology to commercial scale. Although there are many options when it comes to novel methods of sludge disposal and utilization, gasification is currently receiving the most attention. Other novel technologies, such as, super critical water oxidation, which are less mature may be suitable options, but not enough research of these technologies has been performed. With a handful of gasification systems in the final stages of development, only a leap from pilot scale to commercial scale is required, with more technologies following closely behind.

Gasification, when compared to incineration, potentially poses several desirable environmental benefits by reducing and preventing many emissions. One reason for this is the ability to remove compounds through simple cleaning and scrubbing which would later form pollutants during the combustion process. In addition to many possible cleaning methods, gas flow coming from a gasifier is significantly lower than gas flow from an incinerator of the same sludge processing capacity. Typical fixed bed gasifiers require approximately 40 % of the amount of stoichiometric air required for complete oxidation, while incinerators typically require greater than 100 % of stoichiometric air required for complete oxidation (Zainal et al. 2001). The addition of this air in an incineration process increases the total gas flow through the system, requiring larger equipment and handling more dilute gas streams. Many of the CAPs and HAPs are retained in the ash, which is captured in cyclones, removed from the bottom of the gasifier or in bag house filters. This allows better control of the pollutants which can be disposed of properly once consolidated. Pollutants in the gasifier product gas can be captured with water, caustic, or acid scrubbers or dry processes such as zinc oxide, sodium oxide, or calcium oxide beds. Removing these compounds prior to combustion helps to reduce emissions created during the combustion process. Gasification also has the capability of reducing Greenhouse gas (GHG) emissions via utilization of a renewable waste feedstock to produce electricity or thermal energy production which may have been otherwise produced by fossil fuels. The magnitude of GHG reductions is highly site, technology and/or feedstock specific. The destruction of methane during the combustion section of the system and the control of nitrogen during the cleaning process will also help to reduce GHG emissions.

To be socially acceptable, new technology must often prove to be more beneficial in a variety of aspects than traditional practices. Ideally, an overall reduction in

GHG emissions, more favorable HAP and CAP emissions as well as net energy gains, presented in a verified fashion, would quell any social concern. Gasification and incineration processes, convert hydrocarbon-based materials in sludge into simple, nonhazardous byproducts. However, the conversion mechanisms, chemical reactions, and the nature of the byproducts vary considerably (Orr and Maxwell 2000). The clear advantages of gasification are a more versatile product, as syngas may be used in a variety of applications, and lower costs associated with gas cleaning, depending on the ultimate goal, as syngas volume is significantly lower than flue gas volume from an incinerator of a similar processing capacity. The National Energy Technology Laboratory showed the key differences between gasification and incineration as presented in Table 5.3.

5.6.1 Plasma Gasification

Plasma gasification is mainly applied to waste treatment in developed countries. It has great application potential for a wide range of hazardous waste treatment, including both organic and inorganic compounds. Plasma gasification is a gasification process that decomposes biomass into basic components, such as hydrogen, carbon monoxide, and carbon dioxide in an oxygen-starved environment, with the assistance of a plasma torch heating the biomass feedstock to a temperature of 3,000 °C or higher (Rezaiyan and Cheremisinoff 2005). This plasma technique has high destruction and reduction efficiencies.

Mountouris (2008) investigated possibility of plasma gasification for sewage sludge utilization. In general terms, sewage sludge is mainly excess biomass, so it is similar to secondary sludge generated from wastewater treatment at Pulp and Paper plants. Plasma gasification was proven by Mountouris (2008). He reported gasification of 250 tonnes/day sludge. The process was found to be energy efficient (Table 5.4).

The plasma gasification is implemented with the plasma torch, so there is almost no incineration process. The torch reaches temperature level of 3,000 °C that destroys all tars, char, dioxins (Xu and Lancaster 2012). The plasma furnace is two electrodes between which electrical current is passed to generate the electric arc. The gas, usually air, becomes plasma when contacts with the arc. As a result, hydrogen, carbon monoxide and nitrogen are produced mainly. Ash left after the process is vitrified and is very stable and inert so that can be easily applied in the construction materials production. The syngas generated must be purified prior to energy recovery stage, since it still contains acid gases, suspended particles and moisture (Mountouris et al. 2008).

Shie et al. (2014) studied the thermal treatment of wet paper sludge (WPS) and forestry wood waste (FWW) blends (WFB). This process was performed in pilot-scale 10 kW torch plasma and designed to investigate the effects of batch feeding of sample and their results on product yields, gas composition and thermal treatment performance are addressed. From the scanning electron micrograph spectra, the raw

Table 5.3 Comparison of incineration and gasification

Subsystem	Incineration	Gasification
Combustion/gasification	Designed to maximize the conversion of feedstock to carbon dioxide and water	Designed to maximize the conversion of feedstock to carbon monooxide and hydrogen
	Large quantities of excess air	Limited quantities of oxygen
	Highly oxidizing environment	Reducing environment
	Operated at temperatures below the ash melting point	Operated at temperatures above the ash melting point
	Mineral matter converted to bottom ash and fly ash	Mineral matter converted to glassy slag or ash and fine particulate matter (char)
	Gasification	
	Designed to maximize the conversion of feedstock to carbon monooxide and hydrogen	
	Limited quantities of oxygen	
	Reducing environment	
	Operated at temperatures above the ash melting point	
	Mineral matter converted to glassy slag or ash and fine particulate matter (char)	
Gas cleanup	Flue gas cleanup at atmospheric pressure	Treated syngas used for chemical production and/or power production (with subsequent flue gas discharge)
	Treated flue gas discharged to atmosphere	Recovery of reduced sulfur species in the form of a high purity elemental sulfur or sulfuric acid byproduct
	Fuel sulfur converted to SOx during combustion and discharged with flue gas or scrubbed in a flue gas treatment system	
	Gasification	
Residue and ash/slag handling	Bottom ash and fly ash collected, treated, and disposed as hazardous wastes	Slag from a high temperature gasifier is non-leachable, non-hazardous and typically suitable for use in construction materials. Gasifier ash is handled similarly to Incinerator ash
		Fine particulate matter recycled to gasifier or processed for metals reclamation

Based on US EPA (2012)

Table 5.4 Composition of Sewage sludge and heating value in Psittalia

	Dry ash free, %	Dry basis, %	As received, %
Carbon	54.8	37.6	12.0
Hydrogen	8.0	5.5	1.8
Oxygen	33.4	22.9	7.3
Nitrogen	3.8	2.6	0.8
Sulphur	0	0	0
Moisture content	–	–	68 (1)
Ash	–	31.4	10.1
Sum	100	100	100
HHV (MJ/kg)	24.2	16.6	5.3

Based on Deviatkin (2013), Mountouris et al. (2008)

WFB is displaced as long fiber and the construction is complete, however, it become to broken piece after the plasma thermal treatment with ash and small piece of fiber co-existed. Controlled at 873 K of torch plasma reactor, the higher heating value of residue increased to 1.26 time of sample and its maximum value reached to 5,288 kcal/kg. The production of syngas (carbon monooxide and hydrogen) is the major component, and almost 90 % of the gaseous products appear in 2 min of reaction time, with relatively high reaction rates. The maximum instantaneous concentrations and the corresponding time of carbon monooxide and hydrogen occur at 195,255 and 227,950 ppmv, respectively, and 0.75 min for 873 K, with 0.5 min sampling interval. For batch operation, the total syngas yield is about 34.32 wt. % (carbon monooxide of 31.58 and hydrogen of 2.74 wt.%) of raw sample, and the mass ratio of residue is 25 wt.%. The residue from the torch plasma thermal treatment is with the inorganic components converted into non-leachable vitrified lava, which is non-hazardous.

5.6.2 *Supercritical Water Gasification*

With Supercritical water gasification (SCWG) method, biomass can be fully converted into syngas utilising the properties of water at supercritical conditions (Myréen et al. 2010). The research in this area has boomed in recent years. In water at normal conditions there is a significant difference between the liquid and the vapour phase. Supercritical water gasification (SCWG) is based on different physical and chemical properties of water above its critical point (374 °C and 22.1 MPa) (Antal et al. 2000). At these conditions, there is no significant difference in the physical properties between the liquid and vapour phase; there is only one phase, the supercritical phase. Supercritical water acts as, in contrary to water at normal conditions, a non-polar solvent; organic material is hence fully soluble and inorganic material is practically insoluble (Kruse and Dinjus 2007). A large variety of raw material can be gasified in supercritical water; also biomass containing inorganic material is possible to make use of. The products from SCWG are gases, such as hydrogen, carbon monoxide, carbon dioxide, and methane, and product liquid that contain the inorganic part of the raw material and organic material that has not been gasified. The reactions in SCWG are mostly endothermic and constant heating of the reactor is hence required (Kruse and Dinjus 2007). In order to develop a feasible process, efficient heat recovery is required for larger scale applications of SCWG. Up to 70 % of the inserted heat can be recovered from the products; consequently additional heating is required continuously. By using part of the product gas for generation of the additional heat, there is no need for additional heating and the process is in this way self-sufficient (Naqvi et al. 2010). The SCWG method has great potential in being implemented as a waste treatment method for wet waste streams and preferably integrated at the site where the waste material is produced; reducing the need for drying and transportation of the feedstock. The most beneficial side of SCWG process in case of sludge treatment is possibility to feed moist fuels (about 80 weight-% of moisture). Moreover, a small part of hydrogen produced after the process is generated from water itself making hydrogen production more efficient (Gasafi et al. 2008; Boukis et al. 2002). Advantages of Supercritical water gasification are presented in Table 5.5.

Zhang et al. (2010) studied the supercritical water treatment of pulp and paper mill sludge. Sludge of 2 % total solids was tested. However, for higher dryness, more than 25 % total solids is advisable in order to obtain a positive energy balance (Rönnlund et al. 2010). Zhang et al. (2010) studied supercritical water gasification of secondary pulp/paper-mill sludge with an initial water content of 98 wt% in an autoclave reactor at temperatures of 400–550 °C over reaction times ranging between 20 and 120 min. Temperature had a significant effect on improving total gas and hydrogen yields, particularly in the range of 500–550 °C. By contrast, increased reaction times only slightly enhanced gas formation. A notable decline in the total gas and hydrogen yields were observed when the water content was reduced through evaporation of water from the original sludge feedstock. For comparison, supercritical water gasification of three types of sewage sludge (primary sewage sludge, secondary sewage sludge, and digested sewage sludge) from the local municipal wastewater treatment plant was performed under similar experimental

Table 5.5 Advantages of supercritical water gasification

Utilization of different kinds of biomass as an energy source
Suitable for efficient processing of biomass with high moisture content
Complete gasification can be achieved within a short reaction time depending on feed composition
The formation of tar and char depends on feed, reactor design and catalysts
Product gas is available at high pressure in a single step process. This avoids the cost of expensive gas compression
High energy conversion efficiency is achieved by avoiding the process of drying step
Selectivity towards methane, hydrogen, or syngas can be steered with temperature, pressure and using proper catalysts (www.utwente.nl)

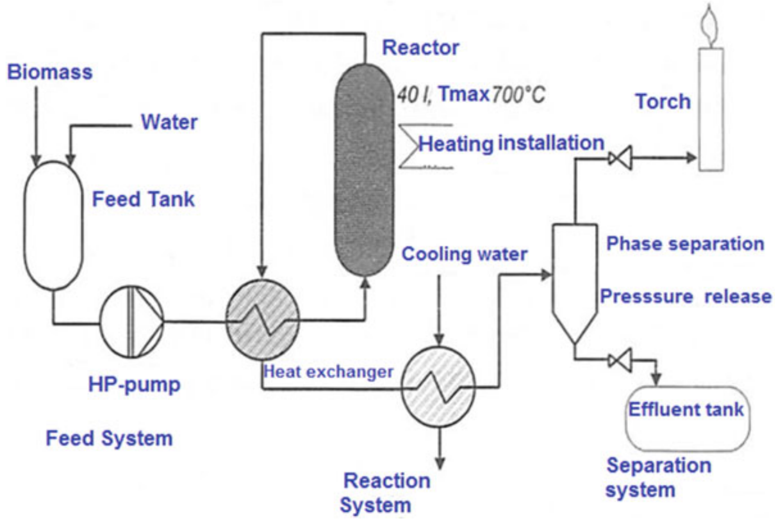


Fig. 5.11 Flowsheet of the Verena pilot plant (Based on Deviatkin 2013, Boukis et al. 2002)

conditions. Secondary pulp/paper-mill sludge was found to exhibit the highest gas product yield, which could likely be attributed to its relatively high volatile matter and alkali metal contents.

Boukis et al. (2002) has provided a description of the process and a simplified flowsheet of a “Verena” pilot plant. The capacity of this plant is 100 t/h (Fig. 5.11). Firstly, sludge is directed into the high pressure pump where it is pressurized up to 30 MPa. Also, the conditioned sludge is heated to 500–700 °C in the gasification reactor. Due to special properties of supercritical water, gaseous components become soluble in water and leave the reactor as a homogeneous phase. Then, effluent from the reactor is cooled down inorder to enable gaseous components to be separated from liquid. In the pilot plant, wastewater generated is only treated from suspended solids and further discharged into a sewage system. In case of deinking sludge, application to suspended solids left after the process must be found to make use of valuable inorganic compounds contained in sludge.

Myreen et al. (2011) investigated two possibilities of integrating a SCWG plant into pulp and paper production. The results are based on mass and energy calculations for a pulp and paper mill and laboratory experiments using black liquor and paper sludge as feed. The experiments were conducted at temperatures of 600–700 °C and pressure of 25 MPa. The integration of a gasification plant in an existing pulp and paper mill will facilitate production of more valuable products than the heat that is generated today. An integration of a SCWG plant would facilitate a development of a traditional pulp and paper mill into a modern-day biorefinery. Future research in the field of recycling of chemicals in the pulping process is essential in design of the industrial scale SCWG plant. The recycling of chemicals is an important issue for the size of the integrated plant. The reactor material and construction of the gasification plant is also a very important issue. More research needs to be conducted, before an industrially applied SCWG process can be built a cost efficient way.

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Chapter 6
Options for Utilization of Waste

Abstract Utilization of Pulp and Paper mill solid waste for Landfilling, Land application (Composting), Vermicomposting, Incineration, Recovery of Raw Materials, Production of Ethanol, Production of Lactic acid, Production of Animal Feed, Pelletization of Sludge, Anaerobic digestion, Paper and Board industry (Fiberboard products, Moulded pulp, Millboard, Softboard), Mineral Based Products (Cement and cementitious products Cement mortar Pozzolanic material, Concrete Ceramic material Lightweight aggregates, Plasterboard Insulating material), Wood adhesive, Sorbent production, Filler in nylon biocomposite production, Landfill Cover Barrier, Bacterial Cellulose and Enzymes, Cellulose-based specialty products, Nanocomposites, Paving and fibrous road surfacing additives are presented.

Keywords Solid waste • Landfilling • Land application (composting) • Vermicomposting • Incineration • Ethanol • Lactic acid • Animal feed • Pelletization of sludge • Anaerobic digestion • Paper and board industry • Wood adhesive, sorbent production, landfill cover barrier • Specialty products • Nanocomposites

Several options are available for utilization of waste from Pulp and Paper Industry. The most utilized methods of pulp and paper sludge management have been land disposal, land application and incineration (Kay 2002, 2003, 2007; CEPI 2004; Gagnon and Haney 2005; Scott and Smith 1995; NCASI 1984, 1992, 1993, 1999; European commission 2006; Spinosa 2004; Spinosa et al. 1994; Valette 1982; Engel and Moore 1998; Glowacki 1994; Garg 2009; Kraft and Orender 1993; Kyllönen et al. 1988; Miner and Gellman 1988; Diehn and Zuercher 1990; Zimmie and Quiroz 1999; Unwin 2000; Watson and Hoitink 1985a, b; Shimek et al. 1988; Bellamy 1995; Bruce 1994; James and Kane 1991; Modell 1985; Ouadi et al. 2012; Frederick et al. 1996; Haintz 2005; James and Kane 1991; Soderhjelm 1976; Haataja and Lund 1980, 1981; Nickull et al. 1991; Coda et al. 2002; Xu and Lancaster 2008, 2009; Cheremisinoff 2002; Mladenov and Pelovski 2010; Porteous 2005; Rodden 1993; Turnbull 1982; United Nations New York 2003; US EPA 2012; Young 1982;

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Garcia et al. 2012; Loatva-Somppi et al. 1998). In 2006, the EPA released a report on biosolids management in the United States which details the current practices, quantities and distribution of sludge disposal methods (US EPA 2006).

6.1 Land Filling

Land filling is the most common method worldwide for the final disposal of paper mill wastes. This situation will not change in many more remote regions of the world in the medium term (Mabee 2001). In contrast, landfill capacities have already become very limited in densely populated countries and it is becoming more and more difficult to obtain authorization to open new landfill sites. Further development of landfilling techniques has dramatically increased costs for construction and operation of landfills (Hamm 2005, 2006). Up to now, approximately 70–75 % of paper mill solid waste has been disposed of in landfill, which is inconvenient from both economical and ecological standpoints (Zule et al. 2007). However, due to decreasing landfill space and new environmental regulations, use of this method will have to be reduced in the future (Gavrilescu 2004, 2005, 2008; Monte et al. 2009; US EPA 1991). Mills have typically favoured landfilling whenever disposal sites are readily available and handling costs are low (Russel and Odendahl 1996). In recent years, however, regulatory agencies have recognized the potential for far-reaching adverse environmental effects from landfilling activities. This has resulted in the tightening of regulations and requirements for more monitoring, environmental impact assessments, closure plans and public consideration. With the current technical state-of-the-art, operating a landfill site such that any annoyance to the population living nearby or environmental risk due to leakage, odors, fire and explosion risk can be largely eliminated. The landfills can be industrial, in that are constructed and operated by the mills, or they can be independently owned, requiring the mills to pay a “tipping fee” for sludge disposal. The European Directive Landfill (1999/31/EC) and upcoming bio-waste directive aims to prevent or reduce, as far as possible, the negative effects of waste landfill on the environment, by introducing stringent technical requirements for wastes and landfill. By increasing landfill fees up to 3.5 €/ton annually until the final landfill use fee will reach 40 €/ton (Likon and Trebše 2012). That fact and adaptation of paper mills to Integrated Pollution Prevention and Control Directive 1996/61/CE forced the paper mill industry to look for processes for their waste minimization and also new technologies for waste reuse or environmental friendly disposal. The major factors to be considered when planning for land fill site include: Environmental suitability of area for land fill; Geology of the area; Environmental impact of run off water from the site; Impact on ground water; Composition and volume of the sludge; Transportation cost. Normal sanitary landfill practices should be observed in constructing an industrial landfill. Theoretically, the following possibilities exist for landfilling waste from paper mills:

- Landfilling in monofill dumps for certain types of waste such as ash, sludge, and bark
- Works-owned landfills where all in-plant waste is stored
- Landfilling in public dumps where waste from other sources is also stored.

About 1 ton of low-ash paper mill sludge in landfill theoretically releases into environment approximately 2.69 tons of carbon dioxide and 0.24 ton of methane (Buswell and Mueller 1952; Likon et al. 2011). According to McKeown (1979) following requirements must be met for landfilling: the disposal site should be a minimum distance above groundwater; the site should be above the flood plain and be protected from flooding; all subsurface conduits-such as culverts, gas and water lines should be removed; the site should be a minimum distance from a public well, highways and watercourse; the nearest property line should be a certain distance away. After a site is chosen, according to the listed criteria, it should be used in accordance with good operating procedures for sanitary landfills (Tchobanoglous 1975).

Several researchers have described the specific requirements for the design of paper mill landfills (Wardwell et al. 1978; Holt 1983; Ledbetter 1976). Modern landfill generally require a liner design. A leachate collection system is required plus FML liners and a clay liner. In daily use, intermediate cover is usually not required, but a final cover will be required, and it must be impermeable, properly sloped, vented, and should have the ability to support vegetation. Most of the environmental effects from landfills arise from the runoff of liquid leached from the waste, that is, the leachate. Leachate is generated at solid waste landfills. This is generated as a result of physical, chemical and biological activity within the landfill. Leachate characteristics are affected by several factors such as precipitation, run-off from and run-on into the landfill, groundwater flow into the landfill, evapotranspiration and consolidation and water generated during the decomposition of the waste. These factors depend on local conditions such as climate, topography, soils, hydogeology, the type of cover on the filled sections, and the type of waste.

NCASI (1992) has reported that the leachates from pulp and paper industry landfills contain:

- Conventional pollutants
- Metals
- Volatile organic compounds
- Phenolic compounds
- Base neutral compounds
- Volatile fatty acids

Metals were usually present at fairly low concentrations (NCASI 1992). Volatile organic compounds were detected; toluene was found to be the most common with a median concentration of 35 μg/L which is well below the Canadian Council of Resource and Environment Minister's goal of 300 μg/L for protection of aquatic life. The only base/neutral compounds found in detectable quantities, more than once were bis-(2 ethyl-hexyl)-pthalate and di-n-octyl pthalate. Pthalates are used in plasticizers, defoamers and lubricating oils. Several kinds of phenolic compounds may be found in pulp mill landfill leachates including cresol isomers, phenols and chlorinated phenols. The decomposition of organic matter under anaerobic conditions produces volatile fatty acids and is common to leachates from many type of landfills. Acetic acid and propionic acid were found in the highest concentrations in pulp and paper mill landfills. A comparison of the average total organic carbon and COD concentrations and the total unsaturated fatty acids (UFA) concentrations showed that UFAs contributed from 7 to 100 % of the organic material in landfill leachetes from kraft mill (NCASI 1992). These leachates if not properly collected and treated may contaminate groundwater or surface water bodies. When landfills are on relatively permeable soils such as sand or gravel, leachate migration may cause contamination over areas many times longer than the area of the landfill. This can also occur over impermeable surfaces such as bed rock where the leachate can flow quickly towards a receptor. Groundwater contamination is a concern if the groundwater is a drinking water source or if it flows to a surface water body. If groundwater contamination directly affects the drinking water supply, the liability implications for the landfill owner/operator may be huge. In addition to impairment of drinking water quality, leachate contamination of ground or surface water may result in the impairment of biological communities, aesthetics and recreational uses. Recognition of these potential effects, together with public awareness of landfilling issues dictates the necessity for a thorough environment impact assessment of new land-fill sites.

In Canada, the regulatory framework does not usually require an environment impact assessment for pulp and paper landfill proposals but, many of the components of an environment impact assessment are fundamental to a successful permitting process. The main components include (Russel and Odendahl 1996):

- Establishing a site development and approval plan,
- Conducting effective public consultation throughout the process
- Undertaking solid technical studies and impact assessment analysis in support of the project

The mill will be required to decide on the specific scope of work based on the environmental conditions of the site, the community needs and the input from local regulatory agencies. Regardless of scope or approach, the mill as a proponent of a new landfill development, must recognize the long-term commitment associated with landfill effects and adopt a management approach which incorporates public involvement with solid technical design and assessment.

Russel and Odendah (1996) have developed a cost effective approach and applied to a landfill in Ontario. Basically, a control chart method is used where for selected leachate indicators, warning and control limits are established. These indicators are selected based on the ratios between background and leachate concentrations, with the highest ratios indicating the most appropriate indicators. The leachate indicators selected should also represent different chemical groups such as metals, nutrients, ions and organic compounds. Before landfill operation, the selected leachate indicators (three to five chemical constituents) are monitored monthly and the concentration differential is used to establish the warning and control limits. The landfill is monitored monthly during the operation and the concentration differential is plotted on a graph for each leachate indicator with the warning and control limits. If the value is within the warning limit, no action is needed, but if the value is above warning or control limits, an established response is implemented to find out the reason and if necessary, initiate control measures. The use of control charts for tracking water quality is advantageous as it is easily interpretable by the public and the mill's environmental managers.

The major drawbacks associated with the land fill is the possible chance of contamination of land and ground water. Because of this reason, most of the developed countries are banning land fill in near future.

6.2 Land Application (Composting)

Composting is an acceptable and recommended means of recycling wastewater treatment sludge and is rapidly gaining acceptance in the world as a method for stabilizing/sanitizing organic wastes. Composting is biooxidative process which involves the mineralization and partial humification of the organic matter; leading to a stabilised final product, free of phytotoxicity and pathogens and with certain humic properties. Several studies of pulp and paper mill sludge composting have been documented (Thacker and Vriesman 1984, 1985, 1986; Drolet and Baril 1997; Macyk 1996; Thacker and Vriesman 1984; US EPA Report 1991; Veillette and Tanguay 1997; Lu et al. 2012; Evanylo et al. 1999, 2000; Gea et al. 2005; Tucker 2005; Baziramakenga et al. 2001; Baziramakenga and Simard 2001; Beauchamp et al. 2002; Bellamy 1995; Bellamy et al. 1995; Brouillette et al. 1996; Charest and Beauchamp 2002; Sesay et al. 1997; Simard et al. 1999; Pepin et al. 1983; Arrouge et al. 1998; Cabral et al. 1998; Camberato et al. 1997; Chantigny et al. 1999, 2000; N'Dayegamiye et al. 2003; Demeyer and Verloo 1999; Glowacki 1994; Goss and Rashid 2004; Koubaa et al. 2010; Matthews 1996; Rantala et al. 1999; Norrie and Fierro 1998; O'Brien et al. 2002, 2003; Jackson et al. 2000; Arrouge et al. 1999; Ekinci et al. 2000, 2002; Macyk and Smith 2000; Marche et al. 2003; Wiseman et al. 2000; Chong 1993; Chong and Cline 1991, 1993; Chong et al. 1987, 1991; Diehn 1991; Thiel 1985; Vasconcelos and Cabral 1993; Zhang et al. 1993; Vagstad et al. 2001; Bockheim et al. 1988a, b; Dolar et al. 1972; Feagley 1994a, b; Chun 2002; Wysong 1976; Mick et al. 1982; Smyser 1982; Carter 1983; Valente et al. 1987; Pridham and Cline 1988; Campbell et al. 1991, 1995; Line 1995; Tripepi et al. 1996; Sesay et al. 1997; Trepanier et al. 1998). The purpose of sludge composting is to biologically stabilize putrescible organics, destroy pathogenic organisms, and reduce the volume of waste. During composting organic material undergoes biological degradation, resulting in a 20–30 % reduction of volatile solids. In composting, aerobic microorganisms convert much of the organic matter into carbon dioxide leaving a relatively stable odor free substance which has some value as a fertilizer. Eccentric micro-organisms are also destroyed due to the rise

Table 6.1 Advantages of composting

The recovery of valuable organic matter and nutrients which are normally lost from agricultural systems when deposited in landfills
Reduced mass and volume of material, and thus reduced transportation costs
Increased concentration of nitrogen and phosphorus through partial mineralisation of the organic fraction, thereby decreasing the risk of damaging plants by the microbial Immobilisation of soil nitrogen
Minimisation of odours which are normally produced following uncontrolled addition of raw organic materials to soils
Transformation of soluble nitrogen and phosphorus into organic forms, thereby extending the availability of these elements over the growing season, and a decreased chance of nutrient leaching
Increased humus content and cation exchange capacity, thereby improving nutrient retention and availability and the production of a higher value product suitable for horticultural and agricultural applications

Based on Finstein and Morris (1975), Shulze (1962), Line (1995), Dick and McCoy (1993), Poincelot (1974), Harada and Inoko (1980), Sesay et al. (1997)

in temperature of the compost. The advantages of composting are presented in Table 6.1.

Composting includes the following operation:

- Mixing dewatered sludge with a bulking agent
- Aerating the compost pile by mechanical turning or the addition of air
- Recovery of the bulking agent
- Further curing and storage
- Final disposal

Composts are not only useful for the improvement of soil quality and crop growth, but can also be used as a substitute for peat in horticultural growing media and as a component in the manufacture of artificial soil. Composts can be used for soil erosion control to reduce plant diseases of agricultural and horticultural crops, act as a substrate for turf and wildflower sods or as a substitute for soil caps on landfills, can assist in the rehabilitation of mine sites and tailings and can be used as biofilters for the immobilisation of odours from industrial processes (Parr et al. 1978; CSIRO Division of Soils 1979; Pagliai et al. 1981; Hernando et al. 1989; Turner 1982; Davey 1953; Duggan 1973; Gouin 1982; Garcia et al. 1991; Shiralipour et al. 1993; Hoitink 1980; Hoitink and Poole 1980; Chen et al. 1988; Fitzpatrick 1989; Hartz et al. 1996; Pinamonti et al. 1997; Poincelot 1974; Weigand and Unwin 1994; Ettlin and Stewart 1993; Daft et al. 1979; Lumsden et al. 1983; Weltzien 1989; Logsdon 1993; Cisar and Snyder 1992; Mitchell et al. 1994; Slivka et al. 1992; Hortenstine and Rothwell 1972; Scanlon et al. 1973; Pinchak et al. 1985; Goldstein 1996).

Aerobic composting is more commonly used than anaerobic composting. The aerobic composting process is exothermic and has been used at the household level as a means of producing hot water for home heating. The major advantage of this compost that it is a very good fertilizer but it is not much used yet.

There has been a lot of interest in use of paper mill biosolids and ink waste in agricultural land for several years. Almost all types of paper industry waste are found to be suitable for composting. These include:

- Fiber-containing sludges
- Deinking sludges
- Bark
- Wood residues
- Biological sludges from effluent treatment plants

Rejects from recovered paper pulping and screening operations are not found to be suitable or suitable only to a limited degree. With this type of waste, the content of plastic and other nonpaper components such as glass or stones has a detrimental effect. In all cases, composting of residues from paper manufacturing requires the application of additives. With the exception of biological sludges, the sludges have an unfavorable carbon/nitrogen ratio for microbial decomposition. They also have a dense structure that is not favorable for composting. This requires the addition of structure-improving materials that are ideally also nitrogen carriers. Biological waste from households, garden waste, cuttings from trees and plants, straw, bark, and waste from animal husbandry are suitable as such components. The composting of fiber sludges and biological sludges, usually with bark, has been practised on an industrial scale for a long time. The paper industry has been using sludges in agriculture for a long time. This is particularly true for virgin fiber sludges and for biological sludges since the first biological effluent treatment plants started operation. With the exception of sludges from biological effluent treatment plants, sludges from the paper industry have a high carbon/nitrogen ratio and therefore contain only a small proportion of nitrogen. For this reason, they have only a limited fertilizing effect. The advantage of their use in agriculture relates to their soil enhancing properties. They contribute to covering the requirements of a humus forming organic substance and also improve the aeration and cultivation of the soil, increase the water retention capacity and prevent erosion.

The composting of paper mill residuals has been done on a commercial scale for about two decades, though only by a small percentage of paper mills. In 1999, some 300,000 tons of residuals were composted by paper mills in United States (Kunzler 2001). Tucker et al. (2004) reported that in UK, composting and vermiculture currently account for just 1,000 tonnes of the residuals produced by the newsprint mills out of the 710,000 fresh weight of sludge produced annually (WRc 2001). Glenn (1997) has listed some of the earlier takers of composting technology:

- Bluestem compost facility which is located in Cedar Rapids, compost sludge from the Cedar River Paper Company. They use yard trimmings and industrial organics as the nitrogen amendment.
- Oneida County in Wisconsin also composts paper residuals which is sourced from Rhinelander Paper. The paper residuals are blended with yard waste.
- In Oswego County, New York, the Solid Waste Authority were driven by a different motivation. They wanted the paper mill sludge as a bulking agent for composting fish and other food processing residuals.

Composting is biooxidative process which involves the mineralization and partial humification of the organic matter. This leads to a stabilised final product, free of phytotoxicity and pathogens and with certain humic properties (Bernal et al. 2009). In composting, process microorganism break down the organic matter of the sludge under aerobic conditions. Traditionally, composting of wastewater treatment sludge has been carried out by using bulking agents. The use of bulking agents are found to enhance the stability of organic matter, inactive pathogens and parasites, and also enable the production of a quality product that may be used as a soil conditioner or as an organic fertilizer (Ponsá et al. 2009; Tremier et al. 2005; Wang et al. 2003; Wei et al. 2001). Several types of materials have been used as bulking agents, although the most widely used materials are wood chips and pruning waste (Atkinson et al. 1996; Larson and McCartney 2000a, b; Wong et al. 1997). The pulp and paper-mill sludge contains significant amounts of plant nutrients and are a widely available resource for composting, thereby curtailing environmental pollution such as direct land applications, reducing landfilling, and limiting greenhouse gas emissions. The compost so produced has the potential to sustain nitrogen-reserves and to improve the structural stability of the soil. It may also be used for horticultural and agricultural applications (Campbell et al. 1997; Hackett et al. 1999). The pulp and paper mills internationally are forced to look for the land application of the sludge as a low cost disposal method due to continued decrease in the availability of landfill space. Land application is found to be suitable both for biosludge and sludge from primary clarifier (Phillips et al. 1997; Sellers and Cook 2003; N'Dayegamiye 2002).

Paper mill sludges have been composted successfully in the past, though both nitrogen and structural amendments are generally required to effect the process. However, traditional composting methods for treating the sludges are limited by their high space requirements. Emerging in vessel technologies for more rapid composting would help alleviate those throughput constraints. Researchers at Ohio State University (Edwards and Burrows 1988) have reported that the quality of the compost product could be increased through a "twin track" process combining composting with vermiculture to produce compost/worm cast blends. As little as 10 % worm cast in a product has shown good benefits with other substrates.

With the diversity of sludges produced, there can be a wide range in the nutrient values amongst sludges (Tables 6.2 and 6.3) (Kunzler 2001). Generally, nitrogen and phosphorus levels are much lower than the levels found in municipal biosolids (sewage sludge), and calcium and magnesium concentrations tend to be higher.

Table 6.2 Nutrient values of paper mill sludges

Material	N (g/kg)	P (g/kg)	K (g/kg)
Primary sludge	0.5–9.0	0.01–40	–
Secondary sludge	6.2–87.5	0.42–16.7	–
Combined primary and secondary sludge	1.1–59.0	0.1–25.4	0.12–10.0
Municipal biosolids	1.0–210	<1.0–150	0.2–650

Based on Kunzler (2001)

Table 6.3 Average parameters of EU paper mill sludges

	Mean values
Dry solids %	32
Carbon/Nitrogen ratio	78
pH	7
Organic matter %	64
N-TK %	1.3
N-NH_4 %	0.02
CaO %	12
MgO %	1
P_2O_5	0.7
K_2O	0.2

Based on WRc (2001)

Table 6.4 Parameters of paper mill sludges relevant to land application

Material	Moisture (%)	C/N	C % dry	N % dry	pH	EC dS/m	Ash % dry
Paper mill sludge	–	–	–	–	7.7	0.2	–
Secondary paper mill sludge	–	41.5	50.6	1.12	–	–	–
Bleached newsprint pulp and paper mill sludge	71.4	218	48	0.22	4.6	0.59	1.64
Paper mill sludge	76.9	218	48	0.22	4.4	0.2	2.1
Mixed deinking, primary and secondary	40–60	89	32	0.36	–	–	49
Primary paper mill sludge	74	–	–	0.3	6.1	–	–
Primary paper mill sludge –bleached kraft/NSSC	–	>168	16.8	<0.1	9.4	–	69.3
Paper mill rejects – bleached kraft/NSSC	–	>357	35.7	<0.1	8.6	–	3.2
Mixed primary, secondary and de-inking sludges	67	48	42	0.87	–	–	25

Based on Kunzler (2001), Chong and Kline (1994), Evanylo and Daniels (1999), Jackson and Line (1997a, b), Sesay et al. (1997), Line (1995), Campbell et al. (1995), Larson and McCartney (2000a), Ekinci et al. (2000)

In general, primary treatment sludges have lower nutrient levels than secondary sludges, particularly with regard to nitrogen. Carbon: Nitrogen ratios also vary greatly amongst sludges (Tables 6.3 and 6.4) and values are generally relatively high, though some secondary sludges can have quite low Carbon: Nitrogen ratios, and some can even be considered as nitrogen sources. Organic solids contents are generally high, though are naturally lower in deinking sludges.

The Waste Management Licensing Regulations, WMLR (SI 1994/1056) in UK, exempt paper mill sludges from requiring a licence for their application to land as a soil conditioner provided that the activity results in a ‘benefit to agriculture or ecological improvement'. Up to 250 tonnes per hectare of exempt waste per annum may be spread on land. This is subject to the provision that the waste provides 'agricultural benefit' or 'ecological improvement' although there is no legal definition of what constitutes a benefit or improvement.

Paper mill sludges for land application for agricultural benefit can originate as primary, secondary, or de-inking sludges, or any combination thereof. Here, distinction needs to be made between the secondary biological sludges which have high moisture contents and show low carbon/nitrogen ratios and the primary and de-inking sludges which are drier but generally show very high carbon/nitrogen ratios as well. It is found that carbon/nitrogen ratios below 17–30:1 will contribute positively to the soil nitrogen balance through nitrogen mineralisation, whilst ratios above 30–35: 1 will act to deplete soil nitrogen (immobilisation), with harmful effects on plant growth.

Extra nitrogen, in excess of the normal plant requirements, must be incorporated with the sludge application to counteract nitrogen immobilisation. But nitrogen immobilisation can be advantageous on certain occasions. Several researchers (e.g. Watson and Hoitink 1985a, b; Honeycutt et al. 1998; Bayer et al. 1992; Zuibilske 1987) have highlighted this issue. Aitken et al. (1995) reported the extra nitrogen requirements to be 40 kg/ha N fertiliser needed for each 100 tonne/ha of paper mill de-inking sludge that was spread. But, on the second year after application, there were no significant effects on crop yield in comparison to a control plot. This showed that the nitrogen was no longer immobilised. Whereas, on plots where a successive, second annual sludge treatment was applied, a continuing nitrogen deficiency was found. In the third year after a single application, there was an overall increase in yield in the treated plot (at 100 tonnes/ha) in comparison to the control. This was due to the increase in the soil organic matter with the sludge treatment.

Nemati et al. (2000) report on the application of paper de-inking sludge as a structural soil amendment. These researchers found that the application did increase structural stability in clay and loamy soils, though in case of a treated sandy loam soil, the structural stability decreased. The effects were again found to be short term. Nemati et al. (2000) considered that an annual sludge application would be needed to sustain any effects. Another soil amendment property, present in many paper mill sludges, lies in their alkaline pH. The potential liming capacity of many paper mill residuals has been noted by WESA and Bates (2002). Overall, in the research trials where paper mill biosolids have been applied to soils there have been few reports of any harmful effects to plant growth if compensatory nitrogen is added alongside. Not much information is available on any potential toxicological effects associated with the land application of paper mill sludges.

The corrugated paperboard division of Menasha Corporation in Michigan USA has been applying mill sludges to land since 1974. With increasing volumes of sludge being produced and a loss of local agricultural outlets through new residential developments, Menasha sought alternative outlets. Here composting played a very important role. Menasha created compost and topsoil products that utilised the mill sludges and woodchip fines in partnership with a composting company (Glenn 1997).

Research on composting of paper mill residuals have generally been conducted in small-scale laboratory reactors. Commercial scale composting operations have also been conducted. Most of the research effort has been to determine the 'best' blends basically through the choice of different nitrogen amendment. A starting carbon:nitrogen ratio of 25–35, in line with the recommendations of Haug (1993) has been targetted. The tested nitrogen amendments have included chicken litter, pig manures and slurries, sewage sludge, paunch together with inorganic sources: ammonium nitrate, urea and other fertilizers (Sesay et al. 1997; Das et al. 1998, 2001, 2002; Ekinci et al. 2000, 2002; Beauchamp et al. 2002; Line 1995; Campbell et al. 1991, 1995; Larson and McCartney 2000a, b; Charest and Beauchamp 2002; Brouillette et al. 1996). Generally, It has been a normal practice to include a structural component to provide porosity and to enable the mixture to be sufficiently aerated. The structural component can be a co-composting substrate, or amendment, like sawdust or wood shavings which degrades along with the paper mill waste, or it can be a coarser bulking agent like wood chips that do not degrade, or degrade more slowly, and are screened out at the end of the composting process for later reuse (Haug 1993). Haug (1993) has reported that the bulking agent does not actually need to be degradable. Poultry manures or pig manures, whilst chosen primarily as a nitrogen amendment, often serve the dual role of a structural amendment also, because of the animal bedding materials that are comingled with the manure.

Norris and Titshall (2011) examined the effects of direct land application of paper mill sludge on plant growth. In a pot trial, application of paper mill sludge to three contrasting soil types at rates of 0, 10, 20 and 40 Mg/ha resulted in an overall decrease in ryegrass yield. Poor plant response, which was generally negatively correlated to sludge application rate, was ascribed to a high sludge carbon: nitrogen ratio and the resulting microbial sequestration of nitrogen, as well as high electrical conductivity and sodium content of the paper mill sludge. Concentrations of phosphorous, calcium, magnesium, sodium and potassium showed variable uptake by the plants with no clear trends evident. While short-term nutrient effects are harmful, the long-term benefits of improved soil physical conditions and increased soil organic carbon should not be discounted.

Aycan and Turan (2014) investigated the effects of different volumetric ratios of bulking agents to pulp/paper-mill sludge on composting. Rice husk and corncob were used as bulking agents. Volumetric ratios of bulking agents to pulp/paper-mill sludge were used as 10:100 and 25:100. To monitor the evolution of the composting systems, routine parameters such as temperature, moisture, pH, total nitrogen, NH4+–N, NO3–N, total carbon, and carbon/nitrogen ratio were analyzed. The results showed that the agro-based materials appreciably affected compost maturity parameters. Furthermore, the quality of the product obtained in the composting process treated with the agro-based materials attained satisfactory stabilization and sanitation for land application.

Several Canadian mills are regularly doing land application and several mills have conducted field trials. QUNO Inc. Thorold, Ontario, Canada has studied land application of primary, secondary and deinking sludges (Pridham and Cline 1988). The characteristics of primary and deinking sludges have been found to have similar. These sludges have low nitrogen and high fiber content. On the other hand, secondary sludges (biosolids) have relatively high nitrogen and phosphorus content and low fiber content. This makes them more suitable for land application. Tests at QUNO showed that the heavy metal content of the combined paper mill sludge was equivalent to that of the cattle manure, and about one-tenth that of municipal sludge. The sludge has been successfully used as a replacement for manure in agricultural applications, and also for land reclamation projects of old sand pits, coal/clinker sites and a former foundry site. Alberta pulp and paper mills worked in conjunction with the Alberta Research Council on land application (Macyk 1999a, b). Land spreading trials were completed on both agricultural and forest cut-block sites. Research was also conducted by the Alberta Newsprint Corporation and Alberta Research Council to evaluate the environmental effect of land spreading conventional and deinking sludge (Pickell and Wunderlich 1995). Preliminary research showed that the procedure should not present any problems in regard to soil quality or plant growth. Trials with land spreading around the mill site have been successfully completed by applying the sludge on top of a gravel base. Alberta Research Council also completed research on ash and sludge land spreading in conjunction with the Slave Lake Pulp Corporation (SLPC) (Pickell and Wunderlich 1995). Grass yield on the test plot site at SLPC showed as much as five times the yield of control plots. SLPC has had favourable results with sludge application on the surrounding agricultural area. Earlier, landfilled sludge was reclaimed and distributed to the farming community and applied using manure spreaders.

In North America, no objections exist concerning the use of deinking sludges in agriculture as a soil improving material provided the material does not exceed defined contaminant concentrations or harmful substance loads. The situation in Germany is quite different. In general, the contaminant concentrations of deinking sludges are well below the valid threshold limits for municipal sewage sludge defined by legislation governing sewage sludge. The German Environmental Protection Agency still considers the agricultural use as unjustifiable for reasons of soil protection, including the irreversibility of numerous soil contaminants. The agency claims the ecological risk potential of deinking sludges is not sufficiently known. In the Biowaste Ordinance issued in 1998, the use of deinking sludges is also not allowable on soil intended for agricultural, forestry, or horticultural purposes.

Domtar Inc. operates a comprehensive land application program for two of its pulp and/or paper mills located in Ontario (Velema 2003). Solid organic residues known as Pulp and Paper Mill Biosolids (PPMB) generated by the effluent treatment plants of both mills are used as soil amendments, fertilizer or mulch in agriculture, silviculture and land rehabilitation projects. The 10-year-old program has developed to sustainably recycle 100 % of PPMB organic type residues that were formerly managed as waste and landfilled. Landfilling and incineration is still a common management option for most of the Pulp and Paper industry in Canada. The Pulp and Paper Research Institute of Canada determined, from the results of a survey in 1995, that industry generated 7.1 million dry tones of residues; 23 % of which were PPMB. An estimated 119,000 dry tones of PPMB were land applied in Canada, representing 7 % of the total available. A 2001 follow-up survey indicated that this had increased to where 42 % practised some degree of land application. Land application of PPMB is safe, ecologically sustainable, environmentally responsible, agronomically beneficial, and economically sensible. Although the regulatory regimes in Canada at the provincial and federal levels discourage the recycling of “industrial wastes”, many opportunities exist for the wise use of these resources through land application, provided programs deal with their communities and publics in an open and proactive manner.

Factors inhibiting the land spreading of many paper mill wastes are their elevated carbon to nitrogen ratios and relatively high Biochemical Oxygen Demands. Supplementary fertiliser additions are essential to prevent nitrogen immobilization and to ensure sufficient nitrogen for crops. Composting pre-treatments will reduce carbon/nitrogen ratios. Composting will also reduce mass, volume and moisture contents benefiting handling, transportation and storage requirements. Overall, the process will produce a stable material, of low odour, with modest levels of nutrients.

Sludges function not as fertilizers but only as amendments. Because they do not contain the elemental analysis required of a fertilizer (Atwell 1981). For a soil amendment, the carbon nitrogen ratio should be 20:1–30:1. An average composition of seven different paper mill combined sludges from ten different mill types was 26:1, so this criteria is being met. The calcium/magnesium ratio should be above 6:1; many combined sludges fail to meet this criteria but the addition of lime to the sludge fulfills it. Sludges are good soil amendments for sandy soils. Detailed analysis of the seven combined sludges did not indicate a heavy metal problem (McGovern et al. 1983). Trials have been conducted in which fly ash and either primary sludge or secondary sludge were applied to crop land. The fly ash-sludge blends were as effective as commercial fertilizer. In these same trials, lime mud applied to agricultural land performed better than dolomite limestone used for the same purpose (Simpson et al. 1983).

Australian Newsprint mills Ltd. (ANM) experimented with biosolids on vegetable and horticultural gardens. The results were found to be encouraging and no detrimental effects were observed (Hoffman et al. 1995). Several farmers have also experimented with biosolids material on small pasture areas and also on orchards, but no objective evaluations were made. ANM carried land spreading trials on crops and pastures because of the high level of interest shown by the farmers (Hoffman et al. 1995). The biosolids were utilized on a farm land close to mill. For this, a desk study and a survey of local farmers was carried out. It was found that biosolids would be readily used by farmers, if it was found to be a viable fertilizer, that it was safe to apply to the environment and the cost was competitive with existing practices. This study also demonstrated that about 2,000 ha per year of land would be required to dispose of the material. It identified the area of interest of land for economic disposal as areas of crops and pasture land within 20 km of the mill and luceme flats where disposal could take place in winter. A field experimental program started with a large area experiment on oats at a location known as Waitara. Biosolids were found to be slow to release their nutrients and produced an effect similar to fertilizer without producing any harmful environmental effects. Rates of 16–64 tonne/ha were required to substitute for normal rates of conventional fertilizer. ANM also carried out trials to spread the biosolids on forest land (Hoffman et al. 1995). Trials started in the Carabost and Green hills State Forests, near Tumbarumba. The major disadvantage with forest spreading over agricultural land spreading is the higher cost of transport to the disposal site. So, the cost of transport would normally make forest spreading unattractive. However, if the solid could be back loaded on log trucks then the economic disadvantage decreases. In Canada, Greater Vancouver Regional District (GVRD) and the University of British Columbia's Forest Sciences department embarked on a 3 year research program at UBC's Malcolm Knapp Research Forest in Maple Ridge to determine the environmental and silvicultural application of recycling pulp and paper sludge and treated sewage sludge as an organic forest fertilizer called Nutrifor (Pickell and Wunderlich 1995). The second phase of the program introduced Nutrifor as a viable fertilizer for forestry and other users.

Scott Paper Ltd. in New Westminster conducted a full-scale land application project with Greater Vancouver Regional District (GVRD). The paper mill sludge was combined with municipal sludge and then applied to a tree farm in the Fraser Valley (Pickell and Wunderlich 1995). In 1990, the GVRD, Western Forest Products Ltd. and the IBEC Aquaculture participated in a fertilization project in which various mixtures of pulp mill wastes, sewage sludge and fish mort silage were applied to forest sites in Southern British Columbia near Port McNeil on Vancouver Island (Taylor et al. 1992). Initial results showed a rapid response by young conifers to organic fertilization. In 1992, a project cosponsored by Nutrifor was completed at Malaspina College where 600 dry tons (2,500 wet tons) of sludge were applied over an area of 26 ha in the Malaspina College Research Forest on Central Vancouver Island (Braman 1993). Full scale projections were made using data obtained from the trials to determine cost per ton of sludge for each of three application methods (Braman 1993). The lowest cost method of spreading the sludge was found to be dry application. Projected cost could be reduced to $ 56/wet ton to apply approximately 36,000 wet tons onto 400 ha.

Seattle, Washington has a sludge management plan which calls for the development of a number of alternative methods (Pridham and Cline 1988). Since halting ocean disposal in 1972, the system has made compost, undertaken strip mine reclamation and is said to have been one of the first to use biosolids in forestry. An innovative application is the growing of hops for the beer industry. Seattle is making use of about 101,000 dry ton/year at 20 % moisture. The effects of lands spreading wastewater sludges from pulp and paper mills were investigated by examining (a) the fate of chlorinated organic materials in landspread sludge and (b) the impact of sludge on plant growth and wild life (Sherman 1995). The results showed that high-molecular-weight chlorolignins were rapidly absorbed by soil or humic matter and organic chlorine was slowly released as inorganic chloride. There was no detectable release of new monomeric chlorolignin-related chlorocompounds. Even under harsh extraction conditions, the extractability of low-molecular-weight chloroaromatic compounds decreased rapidly (half lives of 6–70 days), which was apparently the result of biodegradation and biologically mediated chemical binding into the soil humic structure. No persistent biotransformation products were found. Application of sludges produced an increase in plant growth – grass, hay, corn, trees. Studies of wildlife on sludge-amended soils did not show any harmful effects on the health of individuals or on reproductive parameters. Criteria have also been proposed for the land spreading of solid waste (Springer 1993). The proposed criteria are:

1. The soil sludge mixture must not have a high content of heavy metal that can be taken up by growing plants
2. The soil-waste pH should be 6.5 or higher
3. Excess nitrogen should not be applied beyond that normally taken up by the crop in one season
4. The sludge applied should be free of living pathogenic organisms
5. Solids must be applied in such a manner that they are not available for direct ingestion by domestic animals or humans.

However, land application is not a trouble free technology (Springer 1993). Odours, groundwater contamination, heavy metals, and specific organic toxics are the most common problems. Other problems are noise, pathogens, excessive nitrogen application and surface water contamination. The process of applying sludge is dirty and noisy, so if there are houses in the vicinity, potential difficulties will arise. Actually, public and user acceptance has been very good because sludge is applied mostly to rural areas close to the mill and in some cases on mill owned land.

Pulp and paper mill sludges are usually amenable to well controlled composting methods. Markets for compost include land application for agriculture, horticulture, land reclamation, landscaping and individual consumer use. One mill has had considerable success with marketing its composted sludge. This mill presently composts about 50 % of its sludge. The mill sells the compost to few distributors who market the material in an area within a 250-mill radius from the mill. Initiation of new composting operations within the industry has slowed significantly since the mid-1980s. Lack of sufficiently large, locally available markets for compost and regulatory concerns about the possible presence of chlorinated dioxins and furans in industry sludges are the two common reasons for limited utilization of this management option. Recent industry efforts to reduce the presence of dioxin in sludges are likely to relieve some regulatory concerns about land application of sludges.

One of the mills in United States along with a third party company began to produce synthetic topsoil using sludge (Weigand and Unwin 1994). The process involved the homogenization of sludge with varying proportions of sand, gravel and fertilizer to produce a synthetic soil. More than a dozen landfills have used the soil as part of the final cover. It also has use in other applications requiring vegetative cover. The pulp fiber content of the synthetic soil probably allows for an increased resistance to erosion before the establishment of vegetative cover. In conclusion, composting can add economic value to organic wastes destined for landfill, and use of composts is often associated with a number of environmental benefits.

6.3 Vermicomposting

The solid paper mill sludges derived from the pulp and paper industry are interesting sources of organic matter. On an industrial scale, the pulp mill sludges are usually managed through destructive methods: incineration and landfilling practices (Springer et al. 1996; Tirsch 1990), but vermicomposting could be a suitable technology for its transformation. Vermicomposting is defined as a low cost technology system for the processing or treatment of organic wastes (Hand et al. 1988). Two factors may limit the biooxidation processes: the difficult degradation of the structural polysaccharides and the low nitrogen content of the sludge. These problems could be solved by mixing this waste with some nitrogen-rich materials which act as a natural inoculants of microbial communities. However, the mixture will affect both the earthworm process and the quality of the final product. Therefore, it is very important to know the chemical composition and the best mixture. Scientific investigations have ascertained the viability of using earthworms as a treatment technique for several waste streams (Hand et al. 1988; Raymond et al. 1988; Harris et al. 1990; Logsdon 1994). The action of earthworms in this process is: physical/mechanical and biochemical. The physical/mechanical processes involves substrate aeration, mixing, as well as actual grinding whereas the biochemical process involves decomposition of the substrate by microorganisms in the intestines of the earthworms. These physical/mechanical unit processes usually represent the largest cost associated with a traditional microbial composting process. For that reason, vermicomposting, saves on all these unit operations. Vermicomposting results in the bioconversion of the waste stream into two useful products: the earthworm biomass and the vermicompost in contrast to the traditional microbial waste treatment methods. The earthworm biomass can further be processed into proteins (earthworm meal) or high-grade horticultural compost (Edwards and Niederer 1988; Fisher 1988; Phillips 1988; Sabine 1988) and the vermicompost/castings is also, considered a good product because it has desirable aesthetics, has reduced levels of contaminants, it is homogenous and tends to hold more nutrients over a longer period, without negatively impacting the environment.

Extensive information is available on the vermicomposting of many other materials but not much published information is available on the vermicomposting of paper mill sludges. Just like the composting of paper mill sludges, vermicomposting of paper sludges, needs a supplementary supply of nitrogen. Butt (1993), studied two species of worms fed on a paper mill sludge. Yeast extract was used as a nitrogen source. The earthworms used were Lumbricus terrestris and Octolasion cyaneum. After 120 days none of L. terrestris fed on paper solids alone had begun to develop sexual characteristics, and a mean body mass of only 0.7 g was attained. In the treatments supplemented with yeast extract, mean masses of more than 3 g, were reached within 90 days. Results for O. cyaneum did not follow as clear a trend. Whilst additions of yeast extract enhanced the growth compared with paper only, the responses varied with each yeast. Full maturity was only reached with one of the three yeast feeds. The results showed how earthworm responses to a particular feed can be highly species dependent.

Elvira and Dominguez (1995) conducted study with paper mill sludges from the National Cellulose Company Ltd. in Spain as a feedstock for vermiculture. The sludge, contained 80–85 % moisture, with pH = 9, EC = 0.32 dS/m, nitrogen 0.24 %, carbon:nitrogen 257:1. On its own, the material biodegraded slowly. In its raw state, it did not have lethal effects on earthworms (Eisenia andrei), though the worms suffered weight loss when it was their sole diet. However, when the raw sludge was supplemented by rabbit manure, sewage sludge, or pig or hen slurries, the earthworms grew dramatically. It was considered that an improved nutrient balance and an increase in microorganism populations had both contributed.

Earthworm Eisenia andrei was used by Elvira et al. (1996) to vermicompost paper mill sludge which was mixed 3 parts to 1 with primary sewage sludge. After 40 days vermicomposting, the mean earthworm mass increased from 0.1 to 0.2 g to 0.9 g. All worms developed a clitellum in 21 days and cocoons were produced by 14 days, with the rate of cocoon production increasing fourfold in 40 days. Mortality did not exceed 2 %. Mineralization effects were more pronounced in the worm-worked product than in the control (composted without worms). Regardless of the presence of earthworms, degradation occurred during the bioconversion period, but the presence of earthworms increased the mineralization of organic matter, favored the breakdown of structural polysaccharides and increased the humification rate. As a result, the carbon/nitrogen ratio and the degree of extractability of heavy metals were lower in the worm-worked end product.

Elvira et al. (1997) identified the best source of supplementary nitrogen for vermicomposting the National Cellulose Company sludge. Sewage sludge, pig slurry and poultry manure were all examined. Using Eisenia andrei, the experiment looked at growth and survival in nine mixtures. Increasing the levels of sewage sludge additions resulted in lesser growth and higher mortalities. The paper mill sludge/pig slurry and poultry manure mixtures produced very fast growth, though at the expense of high mortality (more than 25 % survived more than 45 days). Paper mill sludge on its own produced no growth. The maximum final earthworm weight was achieved when paper mill sludge was set at its highest proportion in each respective mix. The mortality in the other treatments was not considered to be due to any lack of food but to the toxicity of the breakdown products of degradation processes and loss of water retention capacity.

Elvira et al. (1998) further studied vermicomposting with Eisenia andrei of sludges from a paper mill mixed with cattle manure in a 6-month pilot-scale experiment. Laboratory experiment was carried out to determine the growth and reproduction rates of earthworms in the different substrates tested. In the pilot scale experiment, the number of earthworms increased between 22- and 36-fold and total biomass increased between 2.2- and 3.9-fold. The vermicomposts were rich in nitrogen and phosphorus and had good structure, low levels of heavy metals, low conductivity, high humic acid contents and good stability and maturity.

These sludges could be potentially useful raw substrates in larger commercial vermicomposting systems, and would reduce the costs related with the exclusive use of different types of farm wastes as feed for earthworms.

Pilot and field scale tests were conducted at the CNR Instituto per la Chimica del Terreno in Pisa, Italy to evaluate the potential of vermicomposting as an economical/environmental alternative in sludge management (Ceccanti and Masciandaro 1999). High quality vermicompost was produced that could be used in the field as a soil organic amendment. A company in Lucca, Tuscany in central Italy planned an industrial plant to process sludges through earthworms.

Research by Butt (1993) had shown that carbon:nitrogen ratios around 25 were the optimum for vermicomposting paper mill sludges, though it was observed that different earthworm species could respond quite differently to different sources of nitrogen. Ndegwa et al. (2000) investigated that carbon:nitrogen ratios of 10:1, 15:1, 20:1, 25:1 based on the total organic carbon and total Kjeldahl nitrogen levels. Substrates were made up from mixtures of sewage sludge and ‘paper mulch’. The experiment lasted 6 weeks with a stocking density of 1.6 kg worms/m^2 and a feed rate of 0.75 kg feed/ kg worms/ day. The worm used was Eisenia fetida. The worm biomass growth decreased with increasing carbon:nitrogen. A carbon:nitrogen ratio of 20:1 or 25:1 showed a net decrease with respect to the initial biomass.

Gajalakshmi et al. (2001) studied four species of worms: Lampito mauritii, Eudrilus eugeniae, Perionyx excavatus and Drawida willsi to vermicompost waste paper. The first two species were found to produce the most worm cast with euginiae also having the better rate of reproduction. Selecting euginiae for further tests, these researchers looked at the effects of worm stocking density on waste paper spiked with cow dung in 4:1, 5:1, and 6:1 ratios. The low-density experiments used 20 worms per 3 l with 75 g supplementary feed, whilst the higher density kept 250 worms per 3 l with 950 g supplementary feed. The results showed that at the low stocking density, 150 mg rising to 220 mg of castings were produced per worm per day. At the higher density, castings production per worm was lower at 100–110 mg/day, but the total weight of castings produced were 6.5 times higher. The proportion of cow dung in the mix had no effect on casting production at the low stocking density though slightly more castings were produced with more dung at the high stocking density. Worm biomass increased more slowly at the higher stocking rate and less food was utilised.

Studies by Dominguez and Edwards (1997) and Frederickson et al. (1997) on earthworm-stocking density have found similar results, with faster growth at lower stocking densities but with highest total biomass production at the highest stocking densities. The optimum feed rates was found to vary greatly not only with earthworm species but also with the feed type (Edwards and Bohlen 1996). Ndegwa et al. (2000) carried out further experiments on stocking densities and feed levels. The results showed that worm biomass increased with stocking density up to 1.6 kg/m^2 then sharply decrease and revealed a weight loss at stocking densities of 2.0 kg/m^2 and above. Biomass increased monotonically with feed rate. Maximum volatile solids reductions also occurred at 1.6 kg/m^2. Volatile solids reductions, however, were not dependent on feed rate.

Ceccanti and Masciandaro (1999) studied mixtures of aerobically-treated sewage sludge and anaerobic paper mill sludge. With 100 % anaerobic sludge, the worms were found to die immediately. Moreover, they did not live for longer than 20 h in the 25 and 50 % anaerobic treatments. To resist the toxic effects, the mixtures were pre-treated through 15 days of daily aeration. Vermicomposting was then carried out for 10 months. A stable product was produced at the end. After 1 month, a 50:50 mix showed 60 % volume reduction and 75 % weight reduction, which resulted from water loss as well as the losses of labile carbon. The 'finished' vermicompost was then matured in windrows (which rose in temperature to 50–60 °C), sanitising the compost through an "accelerated aerobic composting" phase.

The concept of combining composting and vermicomposting to produce an enhanced final product was also considered by Frederickson et al. (1997). The benefits of the combined system would include effective sanitization and pathogen control due to an initial period of thermophilic composting, enhanced rates of stabilisation plus the benefits of vermicompost. Using Eisenia andrei, these researchers compared the straight vermicomposting of green waste with a combined composting, vermicomposting treatment. The growth rate of worms fed on 2-week pre-composted feed was significantly lower than for worms fed on the fresh green waste (reaching 470 mg in 7 weeks compared to 670 mg). Similarly, worms fed on 4-week pre-composted material grew slower than those fed on 2-week pre-composted material, though the difference was not significant statistically. Rates of reproduction in fresh waste and in material pre-composted for 2 and 4 weeks were 1.56, 0.97, and 0.46 cocoons per worm per week, decreasing significantly with increasing periods of pre-composting. Worms fed on fresh waste were all clitellate after 6 weeks whereas those fed material pre-composted for 2 weeks needed 7 weeks to reach maturity. Vermicomposting alone reduced the volatile solids content of the waste faster than composting alone, with the combined treatment showing intermediate results.

Slejska (1996) has made a comparison of the optimum parameters for composting and vermicomposting pulp and paper industry sludges (Table 6.5).

Vermistabilization of paper mill wastewater sludge spiked with cow dung at ratios of 0, 25, 50, 75, and 100 % was carried out by Negi and Suthar (2012) employing the earthworm, Eisenia fetida. A total of five treatments were conducted and changes in chemical and microbial properties of mixtures were observed. Vermistabilization caused decreases in total organic carbon, carbon:nitrogen ratio and cellulose by 1.2–1.5, 4.6–14.6, and 2.3–9.7-fold, respectively, but increases in pH, electrical conductivity, ash content, total nitrogen, available phosphorous, total phosphorous, exchK, calcium, sodium, and N–NO3- of 1.06–1.11, 1.2–1.6, 1.3–1.6, 3.8–11.5, 4.1–6.5, 5.7–10.3, 1.7–2.0, 1.16–1.24, 1.23–1.45, 4.2–13.4-folds, respectively. Paper mill sludge with 25–50 % of cow dung showed the maximum mineralization rate. The fungal, bacterial and actinomycetes population increased 2.5–3.71, 3.13–8.96, and 5.71–9.48-fold, respectively after vermistabilization. The high level of plant-available nutrients indicates the suitability of vermistabilized material for agronomic uses.

Table 6.5 Comparison of composting and vermicomposting methods

Before	Optimal Carbon/Nitrogen	30–35:1	20:1
	pH	6–8	Min. 5; opt. 6.5–7.5; max. 9
	Min. content of P (% of P_2O_5)	0.2	
	Electrolytic conductivity		Max. 3
During	Time of composting (months)	Min. 2–3	Summer: 2–3; winter: 3–5
	Optimal humidity (%)	70	Min. 60; opt. 70–80; max. 90
	Optimal temperature (°C)	Opt. 50–60	Max. 68 min. 5; opt. 18–25; max. 35
	Oxygen demand (% O_2 in environment)		15
	Maximum concentration of CO_2		6
	Maximum height of compost pile (m)	4	0.6 (0.8)
	Maximum content of ammonia (%)		0;1
After	Maximum	30/1	30/1
	pH	6.0–8.5	6.0–8.5
	Humidity (%)	Min. 40; max. 65	Min. 40; max. 65

Based on Slejska (1996)

6.4 Incineration

In the pulp and paper industry there has been a long tradition of waste incineration. It is becoming a more widely used waste management option: burning waste to generate electricity for use at the plant and to sell to the national grid (Xu and Lancaster 2008, 2009; Monte et al. 2009; Kay 2002; Kumar 2000; Loatva-Somppi et al. 1998; Stark et al. 2006; Miner 1981; Oral et al. 2005a, b). Paper sludge has already been used as a fuel source in the Powergen-operated, co-generation plant at Aylesford Newsprint in the UK (Kay 2002). A similar scheme started at the Kemsley site, Sittingbourne, Kent in UK. The solid wastes which are rich in organics are incinerated mainly to reduce its volume and ultimate disposal in a feasible way which is easier and cheaper to land fill. The incinerator generates products of steam and also byproducts of ash and flue gas. The flue gas requires treatment through air pollution control before discharge through the stack. Ash can be disposed directly into the nearest landfill or it can be used as the raw materials for the production of light-weight aggregate and constructional brick for the building industry (Liaw et al. 1998; Monte et al. 2009). However, specifically designed fluidized-bed combustors generate fewer pollutants through the flue gas (Kumar 2000). For power generation, the steam is directed through steam turbines, which work to produce power through the electric generator. Alternatively steam can be used locally for process steam reducing the mill's dependence on costly fossil fuels for steam production (Monte et al. 2009). Typical fluidized bed operating temperature is between 700 and 900 °C (Latva-Somppi et al. 1998). While depending on specific waste regulations in

Europe, a temperature of 850 °C must be achieved for at least 2 s, and with hazardous waste with a content of more than 1 % of halogenated organic substances, the temperature is increased to 1,100 °C for 2 s in order to reduce the formation of the toxic compounds such as polychlorinated dibenzodioxins (Monte et al. 2009).

Sludges and rejects are burned mostly in grate and fluidized bed combustion facilities. Burning of sludges is also conducted in multiple hearth incineration plants. There are several limitation associated with burning of sludge. These are high capital investment, need of auxiliary fuel due to high moisture content, emissions of dioxin, NOx, heavy metals etc. in addition to problems like storage handling, low combustion efficiency and the formation of sticky ash. The sludges are burnt in an incinerator specifically designed for the sludge or in the bark boiler or power boiler. Burning the sludge in the bark boiler, which is a hogged fuel (combination fuel) boiler, seems to cause few problems except for reduced steam generation and reduced boiler efficiency (Miner 1981). A few mills incinerate paper sludge in their boilers as “hog” fuel. This practice is not widespread, because the heating value is very low (Table 6.6) and the high moisture of the sludge affects its ability to burn efficiently.

Incineration in the bark boiler appears to be acceptable for sludge incineration if such a boiler is available on the mill site and if it can take the increased water load. Dewatering to higher levels – 45–50 % solids, will make bark boiler incineration an even more attractive and will reduce the effect on boiler operation.

Combustion properties of a sludge are generally related to the amount of fiber present. Energy available is usually inversely related to the ash content. High ash values up to 50 % on dry basis correlate with relatively low heating values. Sulfur values are important as related to emissions. Dewatering of the sludge stream will be required to increase solids up to some minimum level before combustion will be beneficial or even breakeven. Self-sustained combustion is available with some sludges generated depending on the moisture and organic levels. Cost and benefit evaluations can be made that will indicate the moisture level for optimum performance. Removal of additional water to increase solids above 50 % requires a

Table 6.6 Typical fuel properties of biological and deinking sludge

Heat value and components	Mechanical and biological sludge	Deinking sludge	Peat	Coal	Bark
Heat value – as received, kJ/Kg, (at average moisture)	4,200	2,800	9,200	24,000	5,900
	(63 %)	(58 %)	(50 %)	(12 %)	(60 %)
Ash (dry basis),%	20	50	5	14	3
Elemental analysis (dry basis), %					
C	33.7	19.0	57.1	71.6	50.6
H	4.4	2.4	6.2	4.9	5.9
S	0.3	0.05	0.2	0.6	–
N	0.7	1.0	1.9	1.9	0.5
O	41.2	27.4	29.6	7.0	40.2

Based on Clarke and Guidotti (1995), Gavrilescu (2008) different method similar to paper passing from the press section to the dryer section on a paper machine (Busbin 1995). Thermal drying with hot gases or air can be done in a conveyor dryer, cascade system, or a stand alone drying unit. Reduced water content clearly helps improving the efficiency and also can improve long-term storage options through reduced microbial growth.

The sludge product may be available in several forms depending on the method of combustion and the boiler used. Dewatered sludge straight off a screw press will be lumpy and after moving through several conveying operations begin to break up into a fuel that is fine, uniform and fiberous in nature. To improve handling, storage or combustion characteristics sludge may also be processed further into briquettes or pellets (David 1995; Nichols and Flanders 1995; Sell and McIntosh 1988). Blending dewatered sludge with other fuel (chip fines or saw dust) can help improve conveying characteristics. Pelletizing has come to the forefront as a method to convert combustible solid waste into a usable fuel. Waste to energy via pellet fuels needs to be examined more closely and regarded more highly as a successful solution to landfill crisis. They are quickly becoming a very viable and profitable alternative (Bezigian 1995). Various types of combustion methods are available which include travelling grate boilers, vibrating grate boilers, other hog fuel boilers, bubbling bed combustors, circulating fluidized boilers, stage combustors, rotary kilns, pyrolysis/pulse combustors (Kraft and Orender 1993; King et al. 1994; Fitzpatrick and Seiler 1995). The practicality of the above would be based on the sludge characteristics which are contaminant contents, fuel size, volatility, ash characteristics, heat content etc. and to a great degree the volume to be fired (Busbin 1995). Operating experiences with stoker firing of thermomechanical pulp clarifier sludge with wood waste and combustion of the wastewater clarifier underflow solids in a hog fuel boiler with a new high energy air system have also been reported (King et al. 1994; La Fond et al. 1995). Combined cycle fluidized bed combustion of sludges and other pulp and paper mill wastes to useful energy has been suggested (Davis et al. 1995). Fluidised bed boiler technology provides a means for successful thermal oxidation of high ash, high moisture wastes, producing process steam and/or electricity and reducing at the same time the mill's dependence on costly fossil fuels for steam production (Nickull et al. 1991; Kraft and Orender 1993; Busbin 1995; Fitzpatrick and Seiler 1995; Porteous 2005; Oral et al. 2005a). These systems not only utilize deinking and paper sludges, but also material mined from existing onsite landfills to mitigate ground water contamination problems. Fluidized bed combustion is rapidly becoming the ultimate solution for the final disposal of paper mill wastes (Busbin 1995; Fitzpatrick and Seiler 1995; Davis et al. 1995; Albertson 1999; Porteous 2005; Oral et al. 2005a). It is an emerging technology that works particularly well with the wet sludge produced by the de-inking mills. In this process, air is bubbled through a bed of inert material (usually sand or limestone), which greatly improves the combustion process. This technology also produces fewer sulfur dioxide and nitrous oxide emissions than do conventional hog boilers. Burning sludge is advantageous because the landfill volume required for ash disposal is about 25 % of that required for sludge. In addition, boiler ash from de-inking sludge incineration is sometimes used as an aggregate in cement and concrete. Sludge ash concentrates heavy metals, however, and if their concentration arises hazardous levels, the ash requires special handling (Shin et al. 2005; Usherson 1992). Every tone of recovered fiber generates up to 200 kg (dry weight) of sludge of different types and up to 400 kg (dry weight) of rejects and sludge. De-inking sludge consist of printing inks (black and colored pigments), fillers and coating pigments, fibers, fiber fines, and adhesive components. More than 55 % of the solids removed by flotation are inorganic compounds. They are primarily fillers and coating pigments such as clay and calcium carbonate. The proportion of cellulosic fiber is low. The heating value depends on the ash content and is 4.7–8.6 GJ/t of dry substance (Hamm 2006). The sulfur, fluorine, chlorine, bromine, and iodine contents are low and for this reason, no costly flue gas purification systems are necessary when incinerating de-inking sludge. Compared with sludge from biological effluent treatment plants, the nitrogen and phosphorus contents are very low. This is something that requires consideration when using deinking sludge for composting and agricultural and land application purposes. The level of heavy metals in sludge of recovered paper processing is generally low. Sludge of de-inking plants contains less contamination than those of municipal wastewater treatment. The concentration of cadmium and mercury is especially insignificant and sometimes even below the detection limit of the test method applied (atomic absorption spectrometry). Only the concentration of copper has the same order of magnitude as that of municipal sewage sludge. The copper content of deinking sludge is primarily due to blue pigments of printing inks which contain phthalocyano- compounds (Kiphann 2001).

Pulp and paper companies can improve the cost of operation by using proven, readily available power plant and combustion equipment and systems to efficiently convert the energy available in mill wastes to useful thermal energy and electrical power. By using the combined-cycle concept, either as the combustion turbine combine cycle or the diesel combined cycle, the firing of wood waste and sludge provides net energy gain for the operation of facility rather than merely a means of disposal.

A novel method of thermal treatment of contaminated de-inking sludge has been proposed which is based on the application of the low-high-low temperature (LHL) regions during the combustion (Kozinski et al. 1997). The LHL approach allows for the simultaneous encapsulation of heavy metals within solid particles, removal of submicron particulate and destruction of polycyclic aromatic hydrocarbons before they are emitted into the atmosphere. The encapsulation of the heavy metal layers surrounding the heavy metal rich cores of the ash particles may prevent the metals from leaching under acidic conditions.

Sludge can be easy to burn with the right combustion technology. Knowing that the right technology is very fuel specific and having the technology characterization customized for site-specific conditions is very much important to make proper combustion technology choices. Incineration is not practical for high-ash sludges. Stringent air pollution emissions requirements for combination boilers have diminished the amount of incineration practiced. One of the Finnish mills incinerate sludge if the solids content is over 32 %, and landfills the sludge if it is less than

32 % (Kenny et al. 1995). Operation of the boiler must also be considered when the sludge is not available as a fuel. Several points of consideration include the combustion temperature, fuel feed systems and boiler rating. Older boilers burning sludge as an alternative fuel should be able to simply return to earlier operating states.

Some of the chlorinated organics not eliminated through process modifications could be trapped on the sludge from the external treatment process(es). The disposal of pulp and paper mill sludges, which may contain chlorinated organic compounds, represents an increasing problem. However, if those sludges could be dried to 90 % dry content, in an energy-efficient manner, they could provide high enough flame temperature upon combustion in order to destroy the organic chlorides entrapped in the sludges. In addition, this approach could improve mills' fuel self-sufficiency.

In the Stora Enso Oyj Veitsiluoto Mill at Kemi, Northern Finland, the annual amount of waste to be disposed of in landfill at the pulp and paper mill complex decreased between 1994 and 2008 from 42,990 to 3,197 tonnes (expressed as wet weight). This reduction has been possible through the efficient incineration of burnable waste and by-products (i.e. wood waste and biosludge) in the fluidized bed boiler, used for energy production, together with the effective utilization of waste materials. The ash (ca. 16,860 tonnes; dry weight) originating from energy production in a fluidized bed boiler is reused as a hardener in filling mine cavities (3,301 tonnes; d.w.) at a local mine and in the mill area for landscaping (13,559 tonnes; d.w.).

6.5 Recovery of Raw Materials

Research on the reclamation of fillers from high-ash (deinking) sludges dates to the early 1950s (NCASI 1993; Trutschler 1999; Moss and Kovacs 1995). There are several methods to recover raw materials from sludge (Maxham 1992a, b, c). Paper mill sludges usually contain significant percentages of both cellulose fiber and paper making fillers. Fibre and filler fractionation have been pursued from non-destructive and destructive perspectives, relative to the organic matter components in sludges. The objective of non-destructive material fractionation is to separate usable paper fibre from other solid materials contained in sludge. Early work was focussed on primary sludges, while later work has addressed primary and combined primary-secondary sludge matrices, primarily from virgin fibre or first-run recycle papers. The configuration of the reclamation processes and complexities vary depending on the contaminants present in individual sludges. Work by Maxham (1992b) included the fractionation of secondary fibre and filler substitutes in sludges from deinking operations. Rosenqvist (1978); McAndrew (1985) and Rundell (1985) noted that process implementation and optimization may involve considerable trial and error approaches.

Primary sludges have been the focus of fibre recovery initiatives. In most cases, the approaches to recovering fibres from paper industry sludges involve cleaning and screening processes to separate the usable long fibres from other sludge components. Various system configurations have been employed by various researchers (Rosenqvist 1978; McAndrew 1985; Rundell 1985). Rundell (1985) observed that the reuse of fibre from virgin fibre sludges resulted in increased pitch deposition on mill equipment. This problem could be overcome by returning the reclaimed fibre to the digester blow tank, where black liquor evidently aids in dissolving the pitch. Moss and Johnstone (1993) described results of trials using a Celleco Hedemora Fibre Recovery System and sludge from the Ponderosa Fibre Products mill, Baltimore. Initial sludge analysis indicated that up to 40 % of the sludge matrix consisted of usable fibre (>200 mesh). The system relies on a cleaning stage to remove grit, inks and lightweight contaminants, followed by fibre recovery in a SPRAYDISC filter system. The authors claim good results for both virgin and recycled pulp sludges, although there were no indications of the types of materials to support this claim, other than the work presented in the paper. Full-scale trials were undertaken at the Ponderosa Fibres mill. Results showed the recovery of 20–22 % of the fibre in the sludge, about half of the usable fibre in the sludge matrix. It was evident that additional fibre could have been reclaimed. The operational system at the mill was modified to recover fibre from high-fibre bearing streams before they enter the primary clarifier. It is presumed that this modification was intended to minimize additional sludge cleaning associated with removal of contaminants, grit, dirt and other solids. Prime Fiber Inc. (PFI) constructed a facility to produce market pulp substitute from waste paper and primary paper sludges, with an operating capacity of 30–50 T/day (Ferguson 1992). The PFI process involves the separate pulping, cleaning and screening of the waste paper and sludge streams before mixing. The blended pulp is then processed through a deinking system. Maxham (1992a, b, c) noted that the preferred feedstocks were white paper and primary sludge in which at least 40 % of the matrix consisted of reusable fibre. This, in effect, relies on material containing substantial quantities of virgin fibre materials. The 40 % recoverable level is the minimum level for economic viability. Overall economics of the PFI system have been improved by further developments relative to the addition of filler recovery processes (Maxham 1992a, b). The NCASI (1993) reported one case in which primary sludge from a non- integrated paper mill was assessed as a feedstock for a deinked tissue mill. The project was ultimately abandoned. The product contained unacceptably high dirt levels which were attributed to the sludge additions.

Segregated effluents from paper machines, bleach plants and various cleaning and screening operations can be good targets for fiber reclamation because they usually lack contaminants such as bark or causticizing waste solids. Using some fractionation scheme for the sludge may also provide recovery of fiber alone. The complexity of fiber recovery systems varies widely and depends on the nature of the constituents in the sludge. Mills producing bleached pulp sometimes add recovered fiber to the unbleached pulp entering the bleach plant. This strategy allows for both the reclamation of unbleached fiber and the brightening of previously bleached fiber which may have dirtied by exposure to contaminants in the waste water. Some mills have associated the reuse of fiber recovered from sludge with increased deposits of pitch on equipment. Use of fractionation system helps to recover filler. Most systems for which pilot- or full-scale data are available, have employed a thermal oxidation technique for destroying the organic fraction of the sludge to yield filler in the form of ash (Weigand and Unwin 1994). Experiments with calcination systems have showed that controlling the kiln temperature 816 °C and 843 °C helps to avoid formation of fused agglomerates which can cause the recovered filler to be excessively abrasive. Wet air oxidation method can be also used to recover filler materials from sludge. This process is capable of reducing sludge volume through oxidation of the organic fraction to yield an ash composed of inert materials, e. g., filler clay, titanium dioxide and calcium carbonate for reuse in the papermaking process. One mill in United States is practicing this process on a full scale (Weigand and Unwin 1994). Wet air oxidation is an oxidation reaction carried out in a liquid environment under high temperature and pressure. Initial experience with the operation of WAO unit for filler recovery revealed problems with calcium-sulfate and calcium- oxalate scale deposition. Both pilot- and full-scale systems have demonstrated some problems with low brightness of the recovered filler. In Turkey, primary sludge has been successfully used in the manufacture of hardboard (Ozturk et al. 1992). Full-scale studies using sludge at a 1:4 ratio indicate that the use of 28 bdt/day of waste primary sludge mill save $455,000/year on wood costs and $130,000/year on electricity costs. The method of recovering filler clay from paper mill sludge has been reported (Ellis and Fenchel 1980).

Calcium carbonate and kaolin clay are the two mineral constituents most commonly used in paper making. Calcium carbonate is used in fillers and coating pigments can be further divided into grounded and precipitated calcium carbonate (Omya 2011). Also, talc (magnesium silicates) and titanium oxide can be used. A study by INGEDE showed that calcium carbonate was removed more readily than kaolin in the flotation step, its reduction being two times higher than clay (Göttsching and Pakarinen 2000). Ash of the sludge consists mainly of inert materials, such as clay, titanium dioxide and calcium carbonate. As stated earlier, wet air oxidation method has been used for filler recovery but problems have occurred with brightness of the recovered fillers. The process has been anyway practiced in some mills in the United States to reduce the sludge volume (Weigand and Unwin 1994).

6.6 Production of Ethanol

The idea of converting paper industry sludge to ethanol has been extensively studied in the past decade (Prasetyo and Park 2013; Lark et al. 1997; Ballesteros et al. 2002; Kadar et al. 2004; Yamshita et al. 2008; Marques et al. 2008a, b; Jeffries and Scartman 1999; Peng and Chen 2011). Most of this research has been conducted on sludge emanating from the Kraft pulping process (Fan et al. 2003; Fan and Lynd 2006, 2007a, b; Kang et al. 2010, 2011; Zhang and Lynd 2004, 2010). Some information is also available on the simultaneous saccharification and

Table 6.7 The advantages of paper sludge over other raw materials for ethanol

Paper sludge consists of carbohydrate materials in the form of very fine fibers. These fibres have high specific surface area and lignin is present in very low amount
The sourcing of sludge is easy at practically no cost because it is produced at a concentrated site and permanent production location
The use of sludge for ethanol diverts material going to landfills. This avoids transportation costs and landfill investments

fermentation (SSF) process utilising recycled paper sludge as feedstock. In general, the kraft and sulphite sludge are more amendable to ethanol production than sludge from thermomechanical mills. Deinking sludge is not suitable for bioconversion due to the low content of cellulose fibres associated to the high papermaking process efficiency currently achieved. North Carolina State University (NCSU) researchers have demonstrated that under current market conditions, the process is clearly feasible and profitable. Researchers at NCSU have been developing novel conversion processes to convert paper sludge into ethanol (Gonzalez 2012). The focus has been on low capital investment, operational costs and environmental impact. This project is sponsored by the Biofuels Center of North Carolina and the Consortium for Plant Biotechnology. It is a team effort that involves experts in bioconversion, process economics and financial modeling. The advantages of paper sludge over other raw materials such as agricultural residues or wood sources for ethanol are presented in Table 6.7.

Polysaccharides in recycled paper sludge are much more amenable to enzymatic hydrolysis, in comparison to raw wood or plant material because industrial paper sludge has already been subjected to an extensive mechanical and chemical processing. This process avoids costly pretreatments to make paper industry sludge more accessible to enzymes. It is found to be less energy intensive in comparison to other lignocellulosic biomass-to-ethanol pathways. In fact, the process development of sludge to ethanol via biochemical pathway has been studied over the last few decades. It was found that enzymatic hydrolysis of paper sludge was inefficient in separate hydrolysis and fermentation due to the interference of large amount of ash in the sludges during enzymatic hydrolysis. According to research conducted in the group acid soluble ash like calcium carbonate not only buffers the pH level (usually two to three units higher than the optimum pH) making pH adjustment with acid required for enzymatic hydrolysis, but also adsorbs cellulase with a higher affinity than cellulosic fiber. Acid-insoluble ash like clay also presents inactive binding with cellulase thereby decreasing enzyme digestibility of fiber in sludge. Therefore, in order to achieve higher efficiency in enzymatic hydrolysis with lower enzyme dosage, the researchers at NCSU conducted sludge fractionation before enzymatic hydrolysis in order to separate sludge into two streams: ash-rich stream and fiber-rich streams. This step reduces the acid demand to adjust the pH in enzymatic hydrolysis and also produces an ash-rich stream which can be used for soil amendment. Common pulp washing equipment can be utilized for this fractionation step on an industrial scale. Fractionation step was found to lower the

Table 6.8 Sludge to ethanol production cost share (NCSU Process)

Fractionated sludge-to-ethanol process	Non fractionated process
Enzyme costs of 46 %,	Enzyme costs of 48 %,
Energy costs of 16 %	Energy costs of 13 %
Chemical costs of 6 % of the total production cash cost (production cost minus noncash costs such as depreciation)	Chemical costs of 19 % of the total production cash cost. The non fractionated case has a much higher chemical share of the costs, mainly due to higher sulfuric acid use

Based on Gonzalez (2012)

production cost (including depreciation) to 82 cents per gallon in comparison to \$1.25 per gallon for the process without a fractionation step. The non fractionated process had higher cost in comparison to fractionated process. The chemical cost (mainly sulfuric acid) was sevenfold higher; enzyme costs were 1.8-fold higher and energy costs was 1.5-fold higher (Table 6.8).

Assuming an ethanol wholesale price of \$2.30 per gallon, this process can produce excess returns. The internal rate of return of this process is estimated at 28 % and the modified internal rate of return at 19 % with a reinvestment rate of 8 %. The profitability of the process depends on its simplicity and the business model. This plant is expected to be sited within a paper mill. Therefore, sourcing and buying utilities from the mill reduces capital expenditure. The capital expenditure with this approach is \$4.40 per annual gallon of ethanol and payback of 4.4 years. Paper sludges from different paper making processes are found to perform differently in the technology. Efficiency improvement should be investigated for different types of sludges.

Zhang and Lynd (2010) investigated simultaneous saccharification and co-fermentation (SSCF) of waste paper sludge to ethanol. They used two recombinant xylose-fermenting microbes: Zymomonas mobilis 8b and Saccharomyces cerevisiae RWB222. S. cerevisiae RWB222 produced over 40 g/L ethanol with a yield of 0.39 g ethanol/g carbohydrate on paper sludge at 37 °C, while similar titers and yields were achieved by Z. mobilis 8b at 30 °C. Both S. cerevisiae RWB222 and Z. mobilis 8b exhibited decreasing cell viability at 37 °C when producing over 40 g/L ethanol.

Lin et al. (2012) studied the simultaneous saccharification and fermentation (SSF) process for the biological conversion of pulp and paper sludge into ethanol using Saccharomyces cerevisiae CICC 1001 in batch condition. At the SSF conditions of sterilization, pH 6.0, 6 % of total solid, and adding nutrient solution, the highest ethanol yield of 42.5 g L^{-1} was achieved at the enzyme loading of 40 A.U. (activity unit) g^{-1} VS_{fed} (volatile solid).

Fan et al. (2003), Kang et al. (2010) and Zhang et al. (2009) achieved ethanol concentrations in excess of 40 g L^{-1} using a fed-batch method with paper sludge as feedstock in SSF. In SSCF batch experiments, conversions of paper sludge to ethanol of 51 % were obtained at a solids concentration of 178 g/L (Marques et al. 2008a) with a maximum ethanol concentration of 19 g/L. Ballesteros et al. (2002) attributed low conversion obtained in batch experiments to mixing difficulties at high solid loadings. Mixing is actually required in the reactor to obtain high ethanol conversions. The mixing energy required to mix unreacted paper sludge exponentially increases with solids content. However, when working with a fed-batch method, no substantial increase in mixing energy is required since the low concentrations of paper sludge fed intermittently are hydrolysed which results in a reduction in mixing energy required. The SSF process is normally operated at temperatures ranging between 34 and 37 °C, which is a compromise between the optimal temperatures for the enzymatic hydrolysis of paper sludge and the fermentation of sugars released. The optimum temperature for fermentation by yeast is 30 °C, whereas the optimum temperature for enzymatic hydrolysis is 50 °C (Olofsson et al. 2008). By operating the SSCF process at lower temperatures, inhibition effects on the yeast is reduced as the operating temperature is close to the optimum temperature for the yeast.

Recently, Robus (2013) evaluated nine paper sludge samples obtained from Nampak Tissue Ltd. in terms of ethanol production and those samples yielding the highest and lowest ethanol titres were selected for optimisation. This allowed for the determination of a range of ethanol concentrations and yields, expressed as percentage of the theoretical maximum, which could be expected on an industrial scale. Response surface methodology was used to obtain quadratic mathematical models to determine the effects of solid loading and cellulase dosage on ethanol production and ethanol yield from paper sludge during anoxic fed-batch fermentations using Saccharomyces cerevisiae strain MH1000. This approach was augmented with a multi response optimisation approach incorporating a desirability function to determine the optimal solid loading and cellulase dosage in fed-batch SSF cultures. The multi response optimisation showed that an optimum paper sludge loading of 21 % (w/w) and a cellulase loading of 14.5 FPU g^{-1} be used regardless of the paper sludge sample. The fact that one optimal enzyme dosage and paper sludge loading is possible, regardless the paper sludge feed stock, is attractive since the SSF process can be controlled efficiently, while not requiring process alterations to optimize ethanol production when different batches of paper sludge are processed. At the optimum paper sludge loading and cellulase dosage a minimum ethanol concentration of 47.36 g L^{-1} (84.69 % of theoretical maximum) can be expected regardless of the paper sludge used. The influence of ash on ethanol production was clearly evident since ethanol concentrations from washed samples were up to twofold greater than when unwashed substrate was fermented, irrespective of the enzyme mixture used (Table 6.9). These researchers conducted an economic assessment to ascertain whether ethanol production from paper sludge using SSF was economically viable. It was concluded that paper sludge is an excellent feedstock for ethanol production for the sales of ethanol at a paper sludge feed rate in excess of 50 t day^{-1} with the added environmental benefit of reducing green house gas emissions by 42.5 %.

Recycled paper sludge (RPS) is basically made up of secondary poor-quality non-recyclable paper fibres (fibres too short to be retained on fibre screens and paper machines). The high lignocellulosic content of this sludge material offers therefore an opportunity as feedstock for bio-products (van Wyk and Mohulatsi 2003).

Table 6.9 Ethanol produced from paper sludge in SSF batch cultures with S. cerevisiae MH*

	Ethanol concentration (g/L)	
Enzyme	Washed sludge	Unwashed sludge
Optiflow	5.90 ± 0.44	5.90 ± 0.44
Cellic CTec	3.41 ± 0.16	3.41 ± 0.16
Spezyme	5.09 ± 0.23	5.09 ± 0.23

Based on Robus (2013)

*Standard deviations of triplicate experiments are shown

Moreover, paper sludge is believed to be one of the most promising feedstock for near-term commercial application of technology for converting cellulosic raw materials into commodity products (Fan et al. 2003). In fact, this substrate has some distinctive advantages among cellulosic feedstocks including negative cost at many locations and the potential availability of pre-existing facilities (Fan and Lynd 2007a, b). Marques et al. (2008a) evaluated the possibility of converting recycled paper sludge, an industrial residue stream with strong environmental impact, into valuable products. The approach used was based on the enzymatic conversion of major sludge components (cellulose and xylan) and the simultaneous (SSF) or sequential (SHF) fermentation of the resulting sugars to ethanol. In the enzymatic hydrolysis step using Celluclasts 1.5 L supplemented with Novozyms 188, a degree of saccharification of 100 % was achieved. In relation to ethanol production using the yeast Pichia stipitis CBS 5773, SHF and SSF process efficiencies were compared. A slightly higher conversion yield was attained on SHF, corresponding to an ethanol concentration of 19.6 g L^{-1}, but 179 h were needed. The SSF process was completed after 48 h of incubation allowing the production of 18.6 g L^{-1} of ethanol from 178.6 g L^{-1} of dried RPS, corresponding to an overall conversion yield of 51 % of the available carbohydrates on the initial substrate. These results demonstrate that the biological conversion of sludge to ethanol is efficient even with no pre-treatment or substrate supplementation. Enzymatic hydrolysis yields and major carbohydrates produced from recycled paper sludge are shown in Table 6.10.

Kang et al. (2010) examined two different types of paper mill sludges, primary sludge and recycle sludge, as a feedstock for bioconversion to ethanol. The sludges were first subjected to enzymatic conversion to sugars by commercial cellulase enzymes. The enzymatic conversion was inefficient because of interference by ash in the sludges with the enzymatic reaction. The main reason was that the pH level is dictated by calcium carbonate in ash, which is two units higher than the pH optimum of cellulase. To alleviate this problem, SSCF using cellulase (Spezyme CP) and recombinant Escherichia coli (ATCC-55124), and SSF using cellulase and Saccharomyces cerevisiae (ATCC-200062) were applied to the sludges without any pretreatment. Ethanol yields of 75–81 % of the theoretical maximum were obtained from the SSCF on the basis of total carbohydrates. The yield from the SSF was also found to be in the range of 74–80 % on the basis of glucan. The SSCF and SSF proceeded under stable condition with the pH staying near 5.0, close to the optimum for cellulase. Decrease of pH occurred due to carbonic acid and other

Table 6.10 Enzymatic hydrolysis yields and major carbohydrates produced from recycled paper sludge

				Liberated sugars for hydrolysate (g L^{-1}) DS (%)		
Temperature (°C)	Sludge consistency % (w/v)	Novozyms 188	Incubation time (h)	Glucose xylose	Cellobiose xylobiose	DS (%)
50	3.0	–	144	9.9	1.6	45
				1.9	0.0	
35	3.0	–	72	22.6	1.8	100
				4.9	0.7	
35	3.0	+	72	24.4	0.0	100
				5.6	0.0	
35	7.5	+	120	51.1	6.3	92
				12.0	0.0	

Based on Marques et al. (2008a)

organic acids formed during fermentation. The ash was partially neutralized by the acids produced from the SSCF and SSF and acted as a buffer to stabilize the pH during fermentation. When the SSF and SSCF were operated in fed-batch mode, the ethanol concentration in the broth increased from 25.5 to 32.6 g/L (single feed) to 45 and 42 g/L, respectively. The ethanol concentration was limited by the tolerance of the microorganism in the case of SSCF. The ethanol yield in fed-batch operation decreased to 68 % for SSCF and 70 % for SSF. The high-solids condition in the bioreactor appears to create adverse effects on the cellulase reaction.

6.7 Lactic Acid

Presently, paper sludge is disposed either in landfills or is burnt. If the cellulosic feedstock is utilized for production of useful chemicals, it would not only allow for additional revenues but will also have a positive effect on waste management. Though limited in availability, paper sludge is more promising for such bioconversion processes as compared to other lignocellulosic biomass because it already undergoes processing and thus no pretreatment is required. This makes it an attractive point-of-entry and proving ground for commercial processes featuring enzymatic hydrolysis of cellulose (Yonghong et al. 2008; Fan and Lynd 2006). Many studies have focused on production of different types of value-added chemicals from paper sludge. It has been studied as a substrate for production of carboxy methyl cellulose (Barkalow and Young 1985), activated carbon (Khalili et al. 2000) and for cellulase (Maheshwari et al. 1994). According to Domke et al. (2004), industrial biosludges are a good source of nutrients. It has been widely studied for suitability of conversion to ethanol (Lark et al. 1997; Ballesteros et al. 2002; Kadar et al. 2004;

Table 6.11 Use of lactic acid in food industry

Sodium or potassium lactate in meat, poultry and fish to extend shelf life
Acidity regulator in beverages such as soft drinks and fruit juices
Preservative of vegetables such as olives, gherkins, etc. Preserved in brine
Additive to hard-boiled candy, fruit gums and other confectionery products for reduced stickiness and a longer shelf life
Acidification agent for dairy products which also enhances the dairy flavor
Lactic acid is a natural sourdough acid, which gives the bread its characteristic flavor, and therefore it can be used for direct acidification in the production of sourdough
The esters of lactic acid with long chain alcohols are used as emulsifying agents in bakery products

Yamashita et al. 2008; Marques et al. 2008a; Cheung and Anderson 1996) and lactic acid (Nakasaki et al. 1999; Lee 2005; Marques et al. 2008c). A potential use could be as raw material for lactic acid production. In comparison with alternative feedstocks, Paper mill sludge shows several advantages, including:

- High enzymatic digestibility due to its low lignin content and low particle size
- High protein content
- Negative cost
- Environmental benefit due to the reduction of waste volume

Lactic acid was discovered in 1780 by Carl Wilhelm Scheele, a Swedish chemist who isolated the acid from sour milk as an impure brown syrup. The French scientist Frémy produced lactic acid by fermentation and this gave rise to industrial production in 1881. Pure and anhydrous racemic lactic acid is a white crystalline solid with a low melting point. It exists in two optically isomeric forms, L(+) lactic acid and D(−) lactic acid. L(+) lactic acid is the biological isomer and is ubiquitous in the living kingdom as an important metabolite involved in several biochemical pathways. Lactic acid is an important organic chemical used in several industries. Table 6.11 shows the use of Lactic acid in food industry.

Besides these uses, lactic acid is also an important platform chemical and serves as the precursor of various other useful organic chemicals. For example, by dehydration it yields acrylic acid, oxidation produces malonic acid, while it is hydrogenated to produce 1–3 propanediol. However, over recent years, the biggest surge in demand has been for the polymerization product, polylactic acid which is a biodegradable plastic which has multiple applications in the packaging industry. Two molecules of lactic acid can be dehydrated to lactide, a cyclic lactone, which as biodegradable polyesters is currently used to manufacture tissue engineering materials such as resorbable screws and sutures. It is also increasingly being used as an intermediate in the synthesis of high volume oxygenated chemicals such as propylene glycol, and the esters of lactic acid with low molecular weight alcohols are being used to produce environmental friendly solvents (Dutta and Henry 2006).

Industrially, Lactic acid is manufactured either using a chemical or a biochemical route. The chemical route involves the hydrolysis of lactonitrile using strong acids

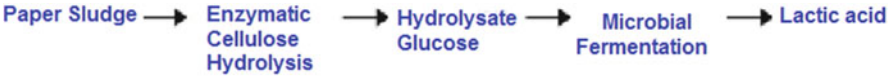

Fig. 6.1 Scheme for production of Lactic acid from paper sludge

that yields a racemic mixture. The biochemical process involves fermentation of sugars such as glucose and lactose (Vadlani et al. 2008) using appropriate microorganisms. Figure 6.1 shows the scheme for production of Lactic acid from paper sludge.

The enzymatic hydrolysis and fermentation steps for lactic acid production can be performed as SHF or SSF. The SSF process offers various advantages over SHF such as the use of a single-reaction vessel for both steps (allowing process integration with the consequent reduction on capital cost), rapid processing time, reduced end-product inhibition of hydrolysis and increased productivity, which is obtained (Sreenath et al. 2001).

Mukhopadhyay (2006) used appropriate cellulase enzymes and efficient microbial cultures and made a comparison of lactic acid production from paper pulp and paper sludge and devised an efficient bioprocess for the conversion of paper sludge into lactic acid. The enzyme requirement for hydrolysis of the cellulose in paper sludge was benchmarked against paper pulp. Enzymatic requirements for complete conversion of cellulose in paper pulp was found to be 12 FPU cellulase, supplemented with 5 EGU of beta-glucosidase per gram of cellulose. However, in the case of paper sludge, beta-glucosidase supplementation had to be increased to 38 EGU to obtain a similar level of hydrolysis indicating a decrease in enzyme activity due to sludge components. Response Surface Methodology (RSM) was used to study the lactic acid yield from paper sludge using enzyme dosage and temperature as parameters and operating in simultaneous saccharification and fermentation (SSF) mode. Maximum lactic acid yield of 0.75 g/g glucose was obtained within 36 h using 10 FPU cellulase supplemented with 32 EGU beta-glucosidase at a temperature of 39 °C. The optimal operational conditions for paper sludge hydrolysis were found to be 9 FPU cellulase, 12.5 EGU beta-glucosidase at 40 °C which resulted in a lactic acid yield of 0.58 g/g glucose using the optimization function of the software. Two Lactic acid producing microbial cultures – Lactobacillus plantarum and Rhizopus oryzae were examined for fermentation of the pulp and sludge hydrolyzate at 125-ml shake flask and 2-L fermenter levels. In paper pulp media, the yields obtained by bacterial and fungal fermentations were 0.89 and 0.36 g/g glucose, respectively. In the case of paper sludge, the yield remained same. However, inhibition of bacterial growth occurred which resulted in lower substrate uptake and productivity in comparison to those obtained in paper pulp. On the other hand, fungal growth rate was increased due to the high solids content of paper sludge. The yield of lactic acid from paper sludge using L. plantarum and R. oryzae was 0.88 and 0.72 g/g glucose, respectively.

Marques et al. (2008c) studied the utilisation of recycled paper sludge as an alternative substrate for lactic acid production. They used Lactobacillus rhamnosus

ATCC 7469, which is reported to provide high productivities and yields. These researchers demonstrated that cellulosic and hemicellulosic fractions of RPS can be completely converted by enzymatic hydrolysis (using Celluclast®1.5 L with Novozym®188) into the constitutive glucose and xylose. These monosaccharides can be used on fermentation media to obtain a variety of products, such as lactic acid, which has an expanding market as precursor of biodegradable polylactides. Maximum production of lactic acid from recycled paper sludge was obtained by performing the hydrolysis and fermentation steps simultaneously on medium supplemented with MRS components and calcium carbonate. L. rhamnosus produced 73 g L^{-1} of lactic acid, corresponding to a maximum productivity of 2.9 g L^{-1} h^{-1}, with 0.97 g LA produced per g of carbohydrates on initial substrate (Table 6.12). A process simplification was also implemented by minimizing recycled paper sludge supplementation and suppressing the addition of Novozym cellobiase.

Romani et al. (2007) conducted a study on the enzymatic hydrolysis of cellulosic biosludges coming from a water treatment plant of a Kraft pulp mill. The effect of the operational conditions such as cellulose to solid ratio, liquid to solid ratio, surfactant concentration and reaction time on the hydrolysates composition was examined, obtaining a set of mathematical models able to predict the glucose and xylose concentrations in the reaction media. Using low cellulase charges (CSR = 8 FPU/g) and high liquid to solid ratios (28–30 g/g), a quantitative conversion of the glucan fraction can be reached in 48 h, but diluted solutions are produced. However, when the operation is carried out using a CSR of 12.5 FPU/g, a LSR of 12 g/g and SC = 0 g/L, 74 % of the glucan fraction and 67 % of the xylan fraction can be saccharified in 34 h, leading to solutions containing up to 27.8 g/L of glucose and 5.4 g/L of xylose. The results demonstrated that this solid residue shows high enzymatic digestibility and that no pretreatments were needed to enhance the saccharification step (detoxification, swelling, etc.). These advantages, along with its negative price, make this solid a valuable raw material for lactic acid production.

Romani et al. (2008) further investigated the use of cellulosic biosludges generated in a Kraft pulp mill as substrate for lactic acid production by SSF. The effect of the operation mode (batch or fed batch), the initial liquid to solid ratio (12 or 30 g/g) and the nutrient supplementation (MRS components or none) on several parameters including lactic acid concentration, volumetric productivity and product yields, were evaluated. When the operation was carried out in fed batch mode with nutrient supplementation and using a LSR0 = 12 g/g, a broth containing 42 g/L was obtained after 48 h with a volumetric productivity of 0.87 g/L h and a product – yield of 37.8 g lactic acid/100 g biosludges. In a similar experiment carried out without nutrient supplementation, a lactic acid concentration of 39.4 g/L was obtained after 48 h with a volumetric productivity of 0.82 g/L h and a product yield of 35.5 g L-lactic acid/100 g biosludges.

Shi et al. (2012) investigated a bioconversion scheme in which the mixture of Paper Mill Sludge and Hemicellulose Prehydrolysate was converted to lactic acid. For this purpose, SSF was applied using Lactobacillus delbrueckii (ATCC 7830) and cellulose enzyme (Novozyme CTec-2). The bioreactor feed was prepared mixing Kraft pulp mill sludge and the prehydrolysate generated from pine wood by

Table 6.12 Lactic acid production with L. rhamnosus

Experiment	F.T.[a]	Cmax[b] (g L^{-1})	S[c] (g L^{-1})	YP/S[d] (g g^{-1})	Y_P/S[e] (g g^{-1})	QP[f] (g h^{-1} L^{-1})	QP max[g] (g h^{-1} L^{-1})
Control fermentation without $CaCO_3$	98	29.8	42.6	0.82	0.70	0.29	0.93
Control fermentation with $CaCO_3$	24	53.9	65.7	0.93	0.82	2.19	4.04
SHF with yeast extract + CaCO3	72	53.7	68.0	0.94	0.79	0.73	2.92
SHF with salts + $CaCO_3$	168	53.1	67.2	0.94	0.79	0.31	2.78
SHF with MRS + CaCO3	168	61.1	67.1	1.09	0.91	0.36	3.03
SHF with $CaCO_3$	168	51.9	72.1	0.91	0.72	0.30	2.59
SSF with neutralised RPS + MRS + $CaCO_3$	168	72.9	–	–	–	0.43	2.86
SSF with neutralised RPS + MRS + $CaCO_3$ without Novozym	168	72.7	–	–	–	0.43	0.76
SSF with non-neutralised RPS + MRS + $CaCO_3$ without Novozym	168	64.9	–	–	–	0.38	0.66
SSF with non-neutralised RPS + MRS without Novozym	168	65.0	–	–	–	0.38	0.66

Based on Marques et al. (2008c)

RPS Recycled paper sludge

[a] F.T.: fermentation time corresponding to maximum lactic acid concentration

[b] Cmax

[c] S: total (glucose, xylose and cellobiose) consumed sugars

[d] YP/S: product yield (lactic acid produced in terms of consumed glucose) calculated at F.T.

[e] YP/S: corrected product yield (lactic acid produced in terms of total consumed sugars) calculated at F.T.

[f] QP: overall lactic acid productivity calculated at F.T.

[g] QP max: maximum lactic acid productivity, which corresponds to the maximal slope in the plot of product concentration versus time

hot-water extraction. The prehydrolysate was detoxified by overliming and charcoal treatment. The SSF was carried out without pH control since calcium carbonate in the sludge, the main inorganic ingredient in the ash, acted as a neutralizing reagent. The main carbohydrate in the prehydrolysate, mannose, and the Kraft sludge were converted efficiently giving above 80 % of theoretical yield with enzyme loading less than 10 FPU/g-hexose. The performance data and the details of the bioprocess, prehydrolysate preparation, and detoxification procedures were reported by these authors.

Tong et al. (2004) studied purification of L(+)-lactic acid from fermentation broth with paper sludge as a cellulosic feedstock using weak anion exchanger Amberlite IRA-92. Some factors such as flow rate, sample volume loaded, pH, and column were systematically examined to improve the purity, yield and productivity in lactic acid purification. Adsorption isotherm of standard lactic acid and lactic acid in the fermentation broth by anion exchanger IRA-92 were also compared. It was observed that in purification process, the increase of pH of the fermentation broth ranging from 5.0 to 6.0 can significantly enhance the recovery yield, purity and productivity. The decrease of flow rate and sample volume loaded can also improve the recovery yield and purity but reduced the productivity. In addition, the scale-up of purification process in laboratory size has little influence on the recovery yield and purity. After optimization, the yield, purity and productivity were found to be about 82.6 %, 96.2 % and 1.16 g LA/(g-resin day), respectively.

Lee (2005) studied production of lactic acid from paper sludge using the SSF process. The SSF process design was based upon the experimental data obtained from cellulose hydrolysis and fermentation. The SSF process was employed to avoid excessively dense solution, when the sludge content in the feed is higher than 15 %, which is one of the several benefits of SSF. In batch SSF, 16 g/L of lactic acid was produced from 5 % paper sludge with an yield of 80 %. Paper sludge which served as a feed appeared to have a buffering effect during SSF, probably due to the inorganic ash component in the sludge. The final product concentration by SSF was observed to be limited by the cellulose content in the system, which can probably be resolved by the intermittent feeding of paper sludge. The SSF of paper sludge via a fed batch mode, with intermittent feeding, produced lactic acid at 162 g/L, with an yield of 74 % and a productivity of 1.4 g/L/h. The modified bioreactor had improved lactic acid production performance after removing indigestible solid materials from the upper compartment, increasing paper sludges feed. For further use of the lactic acid, i.e., polylactic acid, it needs to be recovered and purified. Modeling and simulation for SSF process in bioconversion of paper mill sludge to lactic acid was also carried out. SSF process combined the enzymatic hydrolysis of paper mill sludge into glucose and the fermentation of glucose into lactic acid in one reactor. A mathematical modeling for cellulose hydrolysis was developed, based on the proposed mechanism of cellulose adsorption deactivation. Another model for simple lactic acid fermentation was also developed. A whole mathematical model for SSF was developed by combining the above two models for cellulose hydrolysis and lactic acid fermentation.

Production of lactic acid from paper sludge was studied by Budhavaram and Fan (2009) using thermophilic Bacillus coagulan strains 36D1 and P4-102B. More than 80 % of lactic acid yield and more than 87 % of cellulose conversion were achieved using both strains without any pH control due to the buffering effect of calcium carbonate in paper sludge. The addition of calcium carbonate as the buffering reagent in rich medium increased lactic acid yield but had little effect on cellulose conversion; when lean medium was utilized, the addition of calcium carbonate had little effect on either cellulose conversion or lactic acid yield. Lowering the fermentation temperature lowered lactic acid yield but increased cellulose conversion. Semi-continuous SSCF using medium containing 100 g/L cellulose equivalent paper sludge without pH control was carried out in serum bottles for up to 1,000 h. When rich medium was utilized, the average lactic acid concentrations in steady state for strains 36D1 and P4-102B were 92 g/L and 91.7 g/L, respectively, and lactic acid yields were 77 and 78 %. The average lactic acid concentrations produced using semi-continuous SSCF with lean medium were 77.5 g/L and 77.0 g/L for strains 36D1 and P4-102B, respectively, and lactic acid yields were 72 and 75 %. The productivities at steady state were 0.96 g/L/h and 0.82 g/L/h for both strains in rich medium and lean medium, respectively. The data support that B. coagulan strains 36D1 and P4-102B are promising for converting paper sludge to lactic acid via SSCF.

6.8 Animal Feed

Sludges from pulp and paper mills have been considered promising substrates for single cell protein production (Pamment et al. 1979). There are two basic techniques for using sludges in animal feed. One is to incorporate sludge directly into animal feed mixtures. This method exploits the presence of carbohydrates which are primarily in the form of cellulose and other nutrients present in primary or combined sludges. Research in the early 1970s included experiments on the palatability and digestibility of sludge augmented feed mixtures on goats, sheep, and cattle. The data suggest that the digestibility of sludge relates directly to the carbohydrate content and inversely to the ash and lignin content. Hardwood pulp residues tend to be more digestible than softwood residues (Millet et al. 1973).

The cellulosic component of sludges is available directly to ruminant animals such as cattle, but the digestability is impaired by the presence of lignin and mineral matter. Conversion of mill sludge's cellulosic components to single cell protein can be achieved by a range of bacteria and fungi, but the overall yield is often low (around 25 %) after a high incubation period (about 1 day). The product has been shown to be reasonably digestible, but the economics are very sensitive to the fluctuating price of alternative protein sources. One mill in USA installed a process to convert secondary sludge into a saleable protein product for use in animal feed (Rogers 1982; Evans 1983). Mechanically dewatering secondary sludge to 12 %

solids with further dewatering by feeding a mixture of sludge and oil to specially designed, multiple effect falling film evaporators produced a 45 % protein material. Centrifugation of the evaporator discharge gave 83 % dry solids, 1 % water, and 16 % oil. Targeted markets for the finished product included feed for cattle and poultry and use in agricultural composting (Evans 1983). Unfortunately, acceptance of the product in these markets was not sufficient to support continued production.

Low ash-content primary and secondary sludges were viewed as candidate sources of livestock feed (Harkin 1982; Rogers 1982). High ash-content sludges were deemed to be undesirable due to the intrinsic quantities of inorganic matter, which have no food value, and add bulk only (Harkin 1982).

Pamment et al. (1979) investigated the upgrading of some typical pulp and paper mill solid wastes into protein-enriched animal feed using the cellulolytic fungus Chaetomium cellulolyticum. The waste residues used were six different primary clarifier sludges and a sample of tertiary centricleaner rejects. These were obtained from mills whose modes of operation spanned the range typically in present-day usage: groundwood, sulfite, semichemical, Kraft, and thermomechanical pulping, with and without bleaching. Crude protein production from the solid waste residues is compared to that obtainable from fermentation of untreated or caustic-pretreated sawdusts. Some of these waste residues, especially the Kraft pulp mill rejects, appear to be promising sources of substrate for single –cell protein production. In these preliminary findings, up to 28 % dry weight crude protein content of the product has been obtained at specific growth rates of up to 0.12 h^{-1} on direct utilization of the wastes. Amberg (1984) has reported that Pacific Northwest mill produces more than 18 t/day (20 ton/day) animal feed supplement with a protein content of 45 %. The waste sludge (2 % solids content) is pumped to five belt filter presses for dewatering to a solids content of 12 %. High polymer doses are required for effective dewatering and high solids capture efficiency. The partially dewatered activated sludge is mixed with rendered tallow and pumped to a three-effect falling-film evaporator train for water removal. The patented Carver-Greenfield process is designed to maintain fluidization of solids throughout the evaporation process and fluidization is maintained by mixing six parts of tallow to each part of sludge solids. After water removal, the mixture of oil and biological solids is pumped to a centrifuge and a screw press for oil extraction. The final product is sold as an animal feed and contains about 16 % tallow and 1 % moisture. Although the system successfully dewaters a very difficult sludge, capital and operating costs are substantial and unless a market can be found for the product, the process seems to have limited application in the pulp and paper industry.

ITT Rayonier implemented studies to determine the feasibility of using secondary mill sludge rich in organic matter, nutrients and protein, as a commercial feed product. The results were favourable and resulted in commercial operations. However, the NCASI (1993) reported that this operation was abandoned due to the lack of market acceptance, and poor palatability.

6.9 Pelletization of Sludge

The reasons for producing sludge pellets are:

- Volume reduction
- Odour control
- Recovery of fuel value and
- By-product applications

The most common reason for production of pellets is for use as an alternative fuel. Pellets from waste streams can fuel industrial processes with the energy-equivalence of coal. They can even be used to fuel their own production process. A fully developed system can produce hard, consistent fuel pellets from cellulose waste, and significantly reduce the costs associated with waste-handling. Depending on the composition, 1 ton of pellets can have an energy content as the equivalent energy of coal. The high carbon – content pellets burn efficiently and could fire part of their own production process, thus further reducing costs of production. They can be added to a plant's coal feed. The process can be applied for handling of the waste stream from paper industry. Markets for the pellets have included power plants and cement furnaces. Pellets can be dosed into coal-fired plants of any type, as they are energy-equivalent to coal.

In the United States, one mill transports dewatered, sludge to an off-site pellet mill for drying and formation into pellets. The mill purchases the finished pellets as a fuel supplement (15–20 % water; 10 % ash), having a heating value of 14.7 MJ kg (Kilborn and Weaver 1984; Weigand and Unwin 1994). As the pulp and paper industry is a very intensive user of electricity comprising −12 % of the total manufacturing energy in the United States, significant cost savings may occur through the combustion of sludge (Guidoni 1996).

Einspahr and Fiscu (1984) reported the manufacture of pellets containing dried sludge and a balanced addition of nutrients required for plant growth. In this case, nutrient release occurs slowly over time and may assist in the mineral nutrition of plants. Manufacture of slow nutrient releasing pellets requires specialised machinery. However, if a market for this material could be established, use of sludge in this manner could be a profitable alternative to landfilling. A similar process of sludge recycling has been reported by Anderson (1991). A sludge recovered from a paper deinking mill is converted into a granular product, and is used as a "carrier" material for agricultural and garden pesticides. Apparently the material competes well with other common pesticide carrier materials such as clay or vermiculite, and has the advantage of being dust-free.

Two companies are now manufacturing pellets by using mixtures of sludge and non recyclable paper (Bajpai et al. 1999). These pellets are being marketed as an alternative fuel compatible for use in most stoker and some pulverized coal boilers. The amount of sludge in these pellets can range between 10 and 66 %. It is possible to control the fuel value of the pellets by manipulating both the sludge content and the grade of non-recyclable paper used. The fuel values of the finished pellets are in the range of 14–23 × 10^6 J/kg. The regulatory agencies require evaluation of alternative fuels for by-products of combustion before widespread use of the fuel. Companies involved in both production and use of sludge and NRP fuel pellets have indicated that regulatory reaction to trial run data has generally been positive. NCASI has developed a proprietary process to convert combined sludge from a recovered paper deinking mill into a granular product. The product has been used as a carrier material for agricultural as well as home and garden pesticides and can compete with other common pesticide carrier materials composed of clay, vermiculite, diatomaceous and cob products. Claims for the product indicate that it is superior to some of these conventional carrier because it is dust free and attrition resistant (Weigand and Unwin 1994). The company's production facility has a capacity of 180 tons/day of the granular product.

Kitty littre, poultry littre and large animal bedding have all used pelletized sludge. One mill in USA processes all of its primary sludge into several varieties of animal litter sold to a distributor for marketing. The litter production process is proprietary. It involves sanitizing and deodorizing primary sludge followed by drying and pelletization. Kitty litter is the primary product manufactured, but other products include large animal bedding, pet bedding and bedding for laboratory animals. Grocery stores market kitty litter and feed stores market bedding products. Bedding sells in 25- and 50-lb bags and 1,000 lb tote bins (Weigand and Unwin 1994). Several other companies have studied the feasibility of using sludge to produce kitty or poultry littre. In these cases, they have usually demonstrated production of a quality litter product from primary sludge. Initial capital costs, distribution and marketing issues and incompatibility with company business strategies have inhibited some companies from persuing this byproduct alternative.

6.10 Feedstock for Biochar Production

Biochar is a carbon-rich product. It is produced by thermal decomposition of organic materials (such as wood, manure or leaves) under limited supply of oxygen, and a relative low temperature (700 °C), a process mirror the ancient production of charcoal (Lehmann and Joseph 2009). Use of biochar is not a new concept: the highly fertile terra preta soils in the Amazon have apparently resulted when Amerindians in pre-Columbian times buried charcoal and wastes in the naturally infertile Oxisols (Ferrosols).

The properties of biochar vary widely depending on the feedstock and pyrolysis conditions (Shinogi et al. 2003; Chan et al. 2007, 2008; Gaskin et al. 2008). Biochar is often alkaline, so can have a liming value. Due to its predominantly condensed aromatic structure (McBeath and Smernik 2009) biochar is resistant to chemical and microbial decomposition. Biochars from materials such as manures and papermill sludge have a high content of minerals (Singh et al. 2010), which can be a valuable nutrient source. Research has shown that mean residence time of biochars ranges from hundreds to thousands of years, depending on feedstock and pyrolysis conditions (Singh and Cowie 2008). Manure biochars produced at lower temperatures decompose faster than those made from woody biomass, and at higher temperature.

Biochar can be produced from different biomass sources (Demirbas 2004; Ioannidou and Zabaniotou 2007; Chan et al. 2007, 2008; Chan and Xu 2009; Downie et al. 2009; Lima et al. 2008; Liang et al. 2006). These include:

- Woody materials
- Agricultural wastes such as olive husk, corncob and tea waste
- Green waste
- Animal manures,
- Forestry and crop residues,
- Paper mill sludge
- Poultry waste
- Other waste products

Biochar appears to be a viable option for sequestering carbon in soil (Lehmann and Joseph 2009). Biochar application also possesses potential for enhancing soil fertility which results in increased agricultural productivity (Lehmann and Joseph 2009; Chan et al. 2007, 2008; Steiner et al. 2007). It is also being considered as a soil amendment, adsorbent and fertilizer. It also provides an additional option for carbon sequestration; improving nutrient- and water-use efficiencies (Glaser et al. 2002; Singh et al. 2010). While pyrolysis of a wide range of different biomass and waste materials has been investigated in the past, not much information is available on pyrolysis of paper sludge waste and evaluation of its pyrolysis products.

Conversion of paper mill wastes into biochar through slow pyrolysis has several environmental advantages. LaFleur (1996) reported environmental risks of chemicals used in the conversion of wood to bleached pulp. Papermill wastes can include significant quantities of contaminants including sulphates, fine pulp solids, bleaching chemicals, mercaptans, sulphides, carbonates and hydroxides, casein, clay, ink, dyes, grease, oils and small fibres (Nemerow and Agardy 1998). When applied in the environment, these chemicals have been shown to exert toxic effects in a range of ecotoxicological tests including Pseudomonas putida growth inhibition, Vibrio fisheri luminescence, algal growth, Daphnia magna mobility and Zebra fish hatching and survival (Servos 1996). The thermal processing of wastes into biochar has been identified as an opportunity to destroy contaminants (Glover 2009). This makes beneficial land application possible. According to NCASI (2005) the average paper mill produces around 50 dry kg of paper sludges per tonne of paper produced. These waste sludges are usually disposed of in landfills where they decompose and emit the potent greenhouse gas methane. The importance of eliminating wastes from the paper industry going into landfill has been highlighted in Europe where legislation and increasing taxes are promoting re-use of the wastes through energy recovery projects or land application (Monte et al. 2009). Slow pyrolysis of papermill wastes to produce biochar can potentially achieve the benefits of enhanced plant growth and soil quality seen with biochars from other feedstocks (Chan et al. 2007, 2008).

Most of the early research has basically focused on energy and fuel quality of the biochar product, and the agronomic impacts of biochar have been largely ignored (Home and Williams 1996; Tsai et al. 2006). The impact of biochar on soil properties is likely to vary considerably between different biochars because of the fact that biochar properties are governed by the biomass source and the pyrolysis conditions such as temperature and activation treatment (Chan et al. 2007, 2008; Gaskin et al. 2008; Chan and Xu 2009; Novak et al. 2009; Nguyen et al. 2010).

There are several benefits to be achieved from producing biochar from paper for land application, compared with using the sludge directly as has been proposed by several researchers (Bellamy et al. 1995; Beyer et al. 1997; Phillips et al. 1997). Biochar derived from sludge has improved physical characteristics, such as increased surface areas, and is more friable than the stodgy sludge. The carbon, carbonates and nutrient contained in the sludges are concentrated in the biochar and the product itself is more readily transportable to markets due to being greatly reduced in volume and mass. However, caution should be exercised with flammability of the product.

Biochar produced from paper sludge has been demonstrated to increase productivity in an acidic ferrosol, but had little effect in an alkaline calcarosol (Van Zwieten et al. 2010). Further research is needed before the benefits across a wide range of soil types and crops are determined. It will be possible to undertake cost benefit analysis for the farmers utilising the product. The high content of carbonates in the sludge material, due to the use of calcium carbonate as a whitening agent in the paper making process, means that their use on acidic soils for pH control is advantageous (Boni et al. 2004; Van Zwieten et al. 2010). Direct paper sludge application for acid-mine drainage treatment and in the removal of heavy metals in solution has been suggested by Boni et al. (2004). These functions may be enhanced in the biochar derived from paper sludges. This is due to their increased surface areas and adsorptive properties (Downie et al. 2009) compared with the unprocessed sludge. The greatest uptake of paper sludge derived biochar is to be expected when consumers have commercial quality and environmental assurances related to the product. Possible contaminants in the paper sludge should be assessed for each project application according to the processes used in production. Biochar qualities from each paper sludge source and pyrolysis process should also be reviewed to ensure all risks identified are managed.

Analysis of a primary sludge from one of the Italian mill showed that the sludge does not represent a major threat for the environment in terms of heavy metal release (Boni et al. 2004). If chlorine is used to whiten the paper this could be a potential source of dioxins and furans (Boni et al. 2004), which should be monitored in sludge derived products to be applied to the environment. The net production of greenhouse gases of slow-pyrolysis compared to business- as-usual management of paper sludges needs to be assessed on a case-by-case basis. However the result is likely to be positively affected by the improved resource recovery of the sludge from landfill where a portion of the carbon would be released to the atmosphere as the potent greenhouse gas methane. The stabilization of the carbon into the biochar and the flow-on benefits of biochar application to soil, as discussed in Greenhouse Gas Outcomes, all contribute to a significantly increased greenhouse gas outcome compared to the standard practice for managing paper sludges. It is unlikely, due to paper sludges being very wet, that any energy will remain for export after energy is utilised for internal drying. If however, significant external energy sources from fossil fuels are required to allow the thermal conversion process to progress with this very wet feedstock then any greenhouse gas advantages may be undermined. The energy efficiency of the specific slow-pyrolysis technology will need to be assessed through a complete life cycle assessment to ensure optimised environmental gains are achieved in practice.

Devi and Saroha (2014) performed the risk analysis to study the bioavailability and eco-toxicity of heavy metals in biochar obtained from pyrolysis of sludge of pulp and paper mill effluent treatment plant. The sludge was pyrolyzed at different temperatures (200–700 °C) and the resultant biochar were analyzed for fractionation of heavy metals by sequential extraction procedure. It was observed that all the heavy metals get enriched in biochar matrix after pyrolysis, but the bioavailability and eco-toxicity of the heavy metals in biochar were significantly reduced as the mobile and bioavailable heavy metal fractions were transformed into the relatively stable fractions. Moreover, it was observed that the leaching potential of heavy metals decreased after pyrolysis and the best results were obtained for biochar pyrolyzed at 700 °C. Pyrolysis is a promising sludge treatment method for heavy metals immobilization resulting in significant reduction in the bioavailability and leaching potential of the heavy metals in the biochar. The eco-toxicity of the heavy metals reduced significantly after the pyrolysis, resulting in a decrease in the environmental risk of biochar utilization.

Zwieten et al. (2010) studied amendment of two agricultural soils with two biochars derived from the slow pyrolysis of papermill waste in a glasshouse. Characterisation of both biochars showed high surface area (115 $m^2\ g^{-1}$) and zones of calcium mineral agglomeration. The biochars differed slightly in their liming values (33 % and 29 %), and carbon content (50 % and 52 %). Molar H/C ratios of 0.3 in the biochars suggested aromatic stability. At application rates of 10 $t\ ha^{-1}$ in a ferrosol, both biochars significantly increased pH, CEC, exchangeable Ca and total C, while in a calcarosol both biochars increased C while biochar 2 also increased exchangeable K. Biochars reduced Al availability (ca. 2 cmol (+) kg^{-1} to <0.1 cmol (+) kg^{-1}) in the ferrosol. The analysis of biomass production showed a range of responses, due to both biochar characteristics and soil type. Both biochars significantly increased N uptake in wheat grown in fertiliser amended ferrosol. Concomitant increase in biomass production (250 % times that of control) therefore suggested improved fertiliser use efficiency. Similarly, biochar amendment significantly increased biomass in soybean and radish in the ferrosol with fertiliser. The calcarosol supplemented with fertilizer and biochar gave varied crop responses: Increased soybean biomass, but reduced wheat and radish biomass. No significant effects of biochar were shown in the absence of fertiliser for wheat and soybean, while radish biomass increased significantly. Earthworms showed preference for biochar-amended ferrosol over control soils with no significant difference recorded for the calcarosol. The conversion of papermill wastes to biochar offers industry an attractive option for minimising waste product and reducing costly transport of essentially wet biomass.

Singh et al. (2010) characterised 11 biochars, made from 5 feedstocks [Eucalyptus saligna wood (at 400 and 550 °C both with and without steam activation); E. saligna leaves (at 400 and 550 °C with activation); papermill sludge (at 550 °C with activation); poultry litter and cow manure (each at 400 °C without activation and at 550 °C with activation)] using standard or modified soil chemical procedures. Biochar pH values varied from near neutral to highly alkaline. In general, wood biochars had higher total C, lower ash content, lower total N, P, K, S, Ca, Mg, Al, Na, and Cu contents, and lower potential cation exchange capacity (CEC) and exchangeable cations than the manure-based biochars, and the leaf biochars were generally in-between. Papermill sludge biochar had the highest total and exchangeable Ca, $CaCO_3$ equivalence, total Cu, and potential CEC, and the lowest total and exchangeable K. Water-soluble salts were higher in the manure-based biochars, followed by leaf, papermill sludge, and wood biochars. Total As, Cd, Pb, and polycyclic aromatic hydrocarbons in the biochars were either very low or below detection limits. In general, increase in pyrolysis temperature increased the ash content, pH, and surface basicity and decreased surface acidity. The activation treatment was found to have a little effect on most of the biochar properties. X-ray diffraction analysis showed the presence of whewellite in E. saligna biochars produced at 400 °C, and the whewellite was converted to calcite in biochars produced at 550 °C. Largest amount of calcite was found in Papermill sludge biochar.

6.11 Anaerobic Digestion

Pulp and paper mill sludge has been considered for anaerobic digestion for biogas production (Puhakka et al. 1992b; Ghosh et al. 1975; Ratnieks and Gaylardeb 1997; Weiander 1988). Anaerobic digestion is biological decomposition of biodegradable materials resulting in methane and carbon dioxide production (Bajpai et al. 1999; Bajpai 2000). The microbiology of anaerobic digestion is complicated and involves several bacterial groups forming a complex interdependent food web. However, four major steps can be distinguished. In the first hydrolysis step, both solubilization of insoluble particulate matter and biological decomposition of organic polymers to monomer or dimmers take place. Acidogensis and acetogenesis follow in the second and third step while in the fourth and final step methane and carbon dioxide is produced by methanogenic bacteria (Gavala et al. 2003). The majority of so called biogas produced is methane. The methane in biogas can be burned to produce both heat and electricity, usually with a reciprocating engine or microturbine often in a cogeneration arrangement where the electricity and waste heat generated are used to warm the digesters or to heat buildings. Excess electricity can be sold to suppliers or put into the local grid. Electricity produced by anaerobic digesters is considered to be renewable energy and may attract subsidies. Biogas does not contribute to increasing atmospheric carbon dioxide concentrations because the gas is not released directly into the atmosphere and the carbon dioxide comes from an organic source with a short carbon cycle. The digested and separated solids can undergo further processing and potentially be used as a fertilizer or soil conditioner for land application (Kumar 2000; Gavala et al. 2003), while the treated water may be used for irrigation (Kumar 2000). Anaerobic digestion of municipal or pulp/paper bio-solids could reduce solid wastes by 30–70 % with the benefit of energy recovery through methane production. Generally about half of the organic matter in sludge is susceptible to anaerobic biodegradation into the formation of biogas (Elliot and Mahmood 2007; Mahmood and Elliott 2006). In conventional single-stage anaerobic digestion processes, hydrolysis is regarded as the rate-limiting step in the degradation of complex organic compounds, such as sewage sludge. Ponsá et al. (2008) have proposed a two-stage systems to increase this process The first stage digests the solids, and the second stage separates the undigested solids from the liquid to form carbon dioxide, methane and water. There are two typical operating temperatures for anaerobic digesters determined by the desired species of methanogens. For mesophilic processes, the optimum operating temperature is 37 °C, while 55 °C is desirable for thermophilic processes (Song et al. 2004). Thermophilic anaerobic digestion is generally more efficient in terms of organic matter removal and methane production than the mesophilic process (Gavala et al. 2003). Figure 6.2 shows flow diagram of anaerobic digestion.

Not much information is available related to anaerobic digestion of deinking sludge. Most probably, the treatment of deinking sludge by anaerobic digestion is quite questionable because of its high inorganic content. After all, deinking sludge can possibly contain more toxic compounds that come from inks and other additives used in the printing industry. Anaerobic digestion of WWTS that contains both primary and secondary sludge is not widely used as well, while some

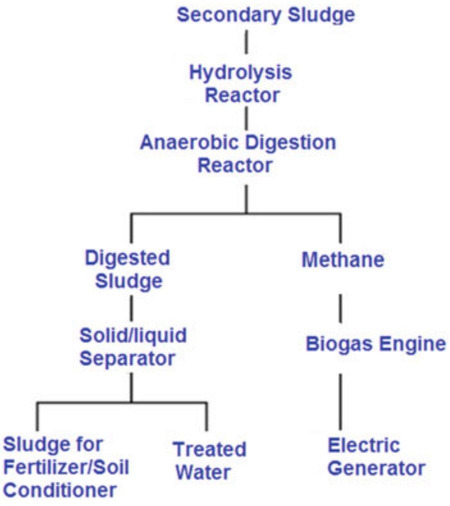

Fig. 6.2 Anaerobic digestion of secondary sludge for biogas production (Based on Kumar 2000; Xu and Lancaster 2009)

installations have been established. The major problem of anaerobic digestion of WWTS generated at Pulp and Paper mills is long residence time of sludge in the digester (20–30 days). However, modern sludge preconditioning methods allow shortening of the residence time to only 7 days. Those pretreatment technologies include:

- Ultrasound treatment,
- Thermal treatment,
- Ozone oxidation
- Mechanical degradation

All methods are aimed at destruction of cellulose walls in order to make them easily degradable (Allan and Talat 2007).

It was found that amount of sludge containing 38 % of lignin can be reduced by 40 %. Resulting biogas production was 0.5 m^3 biogas/kg sludge removed. Optimal performance of the digester was at sludge loading rate of 2.2 kg/m^3 day^{-1}. Produced biogas can be used as fuel for vehicles run of biogas, or in microturbines for electricity production (Talat and Allan 2006). The first successful application of anaerobic digestion for the treatment of pulp and paper mill effluents occurred in the 1970s, having been employed previously for the treatment of sewage sludge for more than 100 years (Rintala and Puhakka 1994). The first anaerobic systems were covered lagoons, but by the 1980s, large, high-rate anaerobic digesters were developed (Rintala and Puhakka 1994). The effectiveness of anaerobic treatment systems for reducing the concentration of organic matter differs between different waste water types. Generally though, significantly less organic matter degradation and methane production occurs in waste waters with a high lignin content, or when resinous acids are present (example tannin and gallotannic acid), as the latter group of compounds are found to inhibit microbial growth (Field et al. 1988). Although anaerobic mixed cultures can decompose monomeric, dimeric and oligomeric lignin model compounds, larger polymeric lignin compounds are resistant to decomposition (Field 1989).

The biomethane potential of Swedish pulp and paper mill sludges has mostly been focused on bio-sludge (Karlsson et al. 2011), whereas the focus in other countries has been mainly on primary sludge (Rintala and Puhakka 1994; Jokela et al. 1997; Lin et al. 2009; Yunqin et al. 2010). The chemical-flocculation sludge has not been investigated separate from the other sludge types as of yet. Nitrogen deficiency is a major concern with anaerobic digestion of pulp and paper mill sludge.

To improve the nutrient status of anaerobic digestion of pulp and paper mill sludge several researchers have studied co-digestion with a nutrient rich waste material. It is considered a low-cost option. Lin et al. (2011) used co-digestion of pulp mill sludge and monosodium glutamate waste liquor to successfully eliminate the nitrogen deficiency problem. Berg et al. (2011) presented a preliminary investigation of anaerobic co-digestion of bio-sludge with a large number of co-substrates, including cows manure. Municipal sewage sludge is an especially suitable co-substrate as it is rich in nitrogen and other required macro and micro nutrients. It is also readily available close to most pulp and paper mills and there is a great deal of accumulated experience of anaerobic digestion operation with this substrate. Furthermore, municipal sewage has been successfully tested in earlier studies of anaerobic digestion of wastewater-treatment sludges from pulp and paper mills (Jokela et al. 1997; Ghosh and Taylor 1999). One of the main advantages of anaerobic digestion is that chlorinated organic substances can be detoxified through reductive dechlorination by microbial consortia (Tiedje et al. 1993; Rintala and Puhakka 1994). Up to 60–87 % of the chemical oxygen demand of a thermo-mechanical pulp mill waste water can be removed by anaerobic treatment (Jurgensen et al. 1985). In chemi-mechanical pulp waste waters, only 40–60 % of the chemical oxygen demand can be removed, due to the presence of compounds which inhibit methanogenesis (Weiander 1988). Depending on the type of sludge or waste water treated, anaerobic treatment can also reduce the volume of sludge by 50–66 % (Takeshita et al. 1981). Since anaerobic digestion does not fully decompose all organic matter present, an aerobic post-treatment process (example, activated sludge treatment) is often required to further degrade compounds which are non-degradable anaerobically (example, resinous acids). After this, the effluent must be clarified to remove the remaining solids, which are then dewatered and transported to a landfill. In some cases, the remaining dewatered sludge is composted and used as a soil conditioner (Rintala and Puhakka 1994).

Anaerobic digestion of Kraft waste activated sludge was investigated in a pilot-scale digester for sludge reduction and biogas production (Puhakka et al. 1992a, b). With sludge containing 38 % lignin, 40 % reduction of the sludge and a biogas production of 0.5 m^3-biogas/kg sludge removed were achieved. In these tests, 13 g NaOH/kg sludge was added to maintain the optimum pH in the system for the maximum sludge reduction efficiency. A Kraft mill in Ontario (Espanola) developed and used an in situ anaerobic fermentation method to digest the primary sludge as opposed to conventional landfilling. The anaerobic process was found to be significantly more economical than the traditional settling basin dredging and land disposal method (Fein et al. 1989). The anaerobic treatment of combined primary and secondary sludge has been shown to produce valuable products. The use of anaerobic digestion has also been reported to pre-treat wastewater, thereby off loading organics entering an aerobic treatment system (Risse and Datschewski 2004; Stahl et al. 2004).

The American–Israeli paper mill in Hadera, Israel, has performed a pilot trial using an anaerobic pretreatment reactor to digest untreated mill effluent (Stahl et al. 2004). The anaerobic digestion of the wastewater resulted in a much lower amount of organics entering the activated sludge system, thus substantially reducing the quantity of waste activated sludge that was required to be wasted from the aerobic treatment system. Stahl et al. (2004) also reported a pilot study using anaerobic digestion to pre-treat wastewater and to digest untreated paper mill effluent, which resulted in a much lower amount of organics entering the activated sludge system, thus significantly reducing the quantity of the secondary sludge from the aerobic treatment operation. Anaerobic digestion has been widely adopted for the treatment of municipal sewage sludge before final disposal and it is employed worldwide as the oldest and most important process for sewage sludge stabilization and treatment (Ponsá et al. 2008). While anaerobic digestion is commonly practiced in the municipal sector, it has not gained popularity in the pulp and paper industry mainly because of its long sludge residence time requirement of 20–30 days (Elliot and Mahmood 2007). There is currently no full-scale anaerobic digestion facility in the pulp and paper sector for the digestion of solid residues. Nevertheless, there is recent technological advancement that potentially can make anaerobic digestion of pulp/paper sludge more feasible by the development and establishment of pretreatment of sludge prior to anaerobic digestion to accelerate the hydrolysis of sludge.

The disadvantage of anaerobic digestion is the high investment and operational costs. These costs can be offset by energy recovered in the form of methane. Both short and long term toxicity problems caused by compounds which inhibit methanogenesis have been associated with some failures in full-scale anaerobic treatment plants (Rintala and Puhakka 1994). Accumulation of large fibre and wood chips in some anaerobic digestion facilities can reduce sludge activity and process efficiency, which requires the reseeding of the digester (MacLean et al. 1990), In general, anaerobic-aerobic systems have been reported to have lower operational costs than aerobic treatment facilities (example, activated sludge processes) because of the reduced aeration-energy requirement, reduced sludge production, and because of methane recovery (Rintala and Puhakka 1994). Thus, widespread treatment of pulp and paper mill effluent or sludge with anaerobic digestion will depend on the relative cost savings incurred-by using the methane recovered, and on the type of material digested.

A Canadian paper mill located in British Columbia has recently installed a high pressure cell disrupter named MicroSludge to breakdown bacteria's tough cell walls of activated sludge followed by anerobic reactor to produce synthetic biogas (Caulfield 2012). Similarly, in a pilot plant study, pulp mill suspended solids was used in an anaerobic bioconversion process to generate biogas at high rates of production (Wood et al. 2009). In fact, biogas synthesis is the only economical viable method to produce biofuels from activated sludges.

6.12 Paper and Board Industry

Several industrial products have been produced from solid waste of pulp and paper mills. The industrial reutilization possibilities can be divided into two categories – ones requiring high inorganic content and a minimum amount of organic compounds, the other ones benefiting from high fiber and low inorganic content of sludge. The most widely researched non-conventional management alternative has been the reuse of primary sludge as feedstock in the manufacture of hardboard (Eroglu and Saatci 1993), fiberboard (Davis et al. 2003; Geng et al. 2006, 2007a, b; Ochoa de Alda and Torrea 2006; Rodland and Helge 1977), building materials such as cement, bricks, concrete (Andreola et al. 2005; Aeslina Abdul Kadir and Mohajerani 2011; Zani et al. 1990; Demir et al. 2005; Dondi et al. 1997a, b;

Cernec et al. 2005; Christmas 2002; Jegatheeswaran and Malathy 2011; Ahmadi and Al-Khaja 2001; Srinivasan et al. 2010; Mohammed and Fang 2011; Vegas et al. 2009; Thomas et al. 1987; Huston 1992; Gallardo and Adajar 2006; Naik et al. 2003, 2004; Naik 2005; Chun 2002; Chun and Naik 2004, 2005; Karam and Gibson 1994), ceramics (Weigand and Unwin 1994), and landfill cover material (Gellman 1989, 1990). These management alternatives upgrade the high fiber content of some primary sludges (example, in fiberboard), the high filler content of others (example, in building materials) or both (example, in landfill cover materials). Several methods to recover fiber and fillers from effluent streams (Wolfer et al. 1997) and primary sludge have been patented (Leuthold and Leuthold 1996; Maxham 1991, 1992a, b, 1994; Simpson and Lam 1998) and implemented (Thermo Fibergen Inc., Bedford, Massachusetts), making possible the reuse of particular sludge components instead of the sludge as a whole. The equipment used to separate fiber or fillers from sludge may be as conventional as screens and cleaners (Dorica and Simandl 1995) or may involve more elaborate methods such as wet-air oxidation (Johnston et al. 2000; Krigstin and Sain 2006).

6.12.1 Fiberboard Products

Some efforts have been made to use paper mill sludge for value-added products, such as gypsum fiberboards or fiberboard additives (Davis et al. 2003). Types of fiberboard include particle board, medium-density fiberboard, and hardboard. Fiberboard is sometimes used as a synonym for particle board, but particle board usually refers to low-density fiberboard (Maloney 1993). In paper mill sludge, and especially in deinking sludge, the amount of intact fibers is small. The sludge, however, has high fines content, the proportion of fines (200 mesh) being 37 % (Davis et al. 2003). The particle size affects the structure, internal bonding and therefore properties of panels produced.

Taramian et al. (2007) used paper mill sludge for particleboard production. Single-layer board and three-layer board, with paper sludge on the surface, were fabricated. Four levels of mixing ratios of paper sludge to wood particles (0:100, 15:85, 30:70, and 45:55) were used. The boards were produced with 3 and 4 % methylene diphenyl diisocyanate (MDI), and 10 and 12 % urea-formaldehyde (UF) adhesives. The bending and shear strengths, water absorption, and thickness swelling of the boards were investigated. The results indicated that the mechanical properties of the produced particleboards are negatively affected by the use of paper sludge (may be due to the inorganic materials present). Overall, UF-bonded particleboards gave superior mechanical performance, water resistance, and thickness swell than MDI-bonded particleboards. The strengths of the UF-bonded board decreased much more than those of MDI-bonded board as paper sludge content increased. The three-layer boards made from 15 % paper sludge with 12 % UF satisfied fully the minimum requirements set by EN, ASTM D 1037-99, and ANSI A208.1 standards for general uses.

Xing et al. (2012) used secondary sludge from three conventional pulping processes -thermomechanical pulp, chemical-thermomechanical pulp and Kraft pulp from three mills. The buffering capacity and pH of secondary sludge and particle of SPF (spruce, pine and fir) were measured. The dry secondary sludge as co-adhesive was added in the formulation of particleboard manufacturing. The urea-formaldehyde content was 7 % (dry weight of resin per dry weight of particle), and the different percentages of secondary sludge (dry weight of secondary sludge per dry weight of particle) were added in the particleboards. Each combination of urea-formaldehyde – secondary sludge type – secondary sludge content was considered as a different formulation, with three replications for each one. The internal bond strength was measured. Results indicated that secondary sludge can be used in particleboard manufacturing and there is an optimal percentage for each secondary sludge type, whereas the value of percentage depends on the type of secondary sludge and paper mill.

Geng et al. (2007a) characterized de-inking paper sludge and primary sludge containing 20 % secondary sludge from a paper mill as to their suitability for the manufacture of medium density fiberboard. Compared with de-inking paper sludge, de-inking paper sludge had a lower ash content, higher holocellulose content, more and longer fibers, lower pH, and higher buffering capacity. These characteristics make paper sludge a better fiber resource for fiberboard than deinking paper sludge. Fiberboards were manufactured at the Pilot Plant of Forintek (Québec City, QC, Canada) using virgin SPF and primary sludge or de-inking paper sludge at different sludge/SPF weight ratios with 12 % urea formaldehyde resin. At an equal sludge/SPF weight ratio, primary sludge-SPF panels had much higher mechanical properties than did deinking paper sludge -SPF panels. At a primary sludge /SPF weight ratio of 7:3, the mechanical properties of paper sludge -SPF panels were higher than the requirements of ANSI A208.2-2002 MDF standard for Grade 120 in terms of internal bond strength, modulus of rupture, modulus of elasticity, and thickness swelling. With deinking paper sludge /SPF weight ratios as low as 3:7, the tested mechanical properties of deinking paper sludge-SPF panels could meet the requirements of ANSI A208.2-2002 MDF standard for Grade 120.

According to Maloney (1993), a board produced from sludge with high fines content and a synthetic resin would be called “fiberboard”. The use of deinking sludge in the production of medium density fiberboard, also known as MDF, has been studied by Geng et al. (2007a) and Davis et al. (2003). Fiberboard panel is one of the most popular materials used in furniture and building applications (Geng et al 2007a). MDF is the most typical fiberboard and is usually used in the furniture industry, wall paneling or e.g. in dashboards and inner doors of cars. The composition of fiberboard can vary greatly. In a study by Davis et al. (2003), phenol formaldehyde resin was used, the rest of dry solids was fiber (or partially clay and calcium carbonate when deinking paper sludge was used). Geng et al. (2007a) used urea-formaldehyde resin with fiber mixtures. A board made from glass and cellulose fibers and synthetic resin could consist of 20 % resin and 80 % fiber of which 25–50 % cellulose, and the rest glass fibers (Bullock 1984). Fiberboard compositions and composition of deinking paper sludge, are presented in Table 6.13.

Table 6.13 Compositions of fiberboards and the corresponding deinking sludge compositions

Fiberboard composition	Davis et al. (2003)	Geng et al. (2007a)
Resin	6 %	12 %
Virgin fiber	47 %	70 %
Deinking paper sludge fines of deinking paper sludge clay + calcium carbonate of deinking paper sludge	47 %	30 %
	35 %	–
	20 + 4 %	54 %

Based on Davis et al. (2003), Geng et al. (2007a)

Davis et al. (2003) investigated the mechanical and physical properties of MDF to which was added deinking paper sludge as an alternative to the current practice of disposal of deinking paper sludge in landfill facilities. A treatment method was developed to reduce the moisture content and consistency of the deinking paper sludge to within a range suitable for the production of MDF (6–9.5 % moisture content). Treated material was found to contain approximately 11 % silicon, 10 % aluminum, 4 % calcium, and 2 % titanium. Thirty-seven percent of the treated material passed a 200 mesh screen (fine content). Panels were produced using virgin MDF fiber (6 % phenol-formaldehyde resin), which included additional factors chosen based on deinking paper sludge composition. These factors (by percent oven dry furnish weight) included fine content (0–35 %), kaolin coating clay (0–20 %) and calcium carbonate from (0 to 4 %). A response surface regression model equation was then generated for each property. Coating clay was the primary factor affecting mechanical properties, exhibiting linearly negative effects on modulus of elasticity, modulus of rupture and internal bond strength. Moisture content, change in conditioned weight, and conditioned volume were found to linearly increase along with clay content. By contrast, the equation for conditioned moisture content was dependent on fine and calcium carbonate content and included quadratic and linear terms. The model equation for the moisture content at 30 % relative humidity in the linear expansion test included a linear calcium carbonate term as well as quadratic terms for both clay and calcium carbonate. Flammability of the model panels was decreased by increasing clay content and was found to have a strong positive correlation with increased fiber content. Decay by the brown-rot fungus G. trabeurn increased with clay content and decreased with increased calcium.

Davis (2003) and Geng et al. (2007a) reported that inorganic content, especially clay, reduces mechanical properties of MDF. Otherwise there seemed to be no physical or operational hinders to prevent deinking paper sludge use as an addition in fiberboard production. In the latter study a fiberboard incorporating deinking paper sludge even managed to meet ANSI standards. A small proportion of inorganic or 35 % proportion of fines did not have significant effect on board properties. This implies that if the filler content were at least partially removed from the sludge, utilization of deinking paper sludge in MDF production could be a viable opportunity. No allowed concentrations or threshold limits of trace element heavy metals in fiberboard were found in literature.

Jesus and Alda (2008) reviewed the fiber content, fiber quality, and key physical and chemical properties (humidity, ash content, abrasiveness, drainability, and oxygen uptake) of 20 different primary sludges sampled in European mills. Although sludge characteristics are highly variable across pulp and paper mill processes, sludges can be considered to fall into two main types:

- High-ash sludge (more than 30 % dry weight)
- Low-ash sludge (less than 30 % dry weight).

Results of paper tests (caliper, breaking length, tear index, elongation, bursting strength, stiffness, opacity, whiteness, and porosity) and board tests (ring crush test, Concora medium test, corrugated crush test) suggest that at least 12 of the sludges studied could be reused in the paper and board industry. The results make it possible to differentiate three primary sludge grades: the first needs little cleaning and has appropriate strength properties to be a component of printing and writing papers, tissues, and wrapping papers; the second requires cleaning, bleaching, or both and has appropriate strength properties for applications that do not require high brightness, such as corrugated board, boxboard, and some tissue grades; the third requires cleaning and has limited strength properties, but could be used in some mills that operate using closed water cycles because the final product can tolerate a certain degree of dirt and contamination, as in some packaging and construction-paper grades. Primary sludges share several features in common with recycled paper, and therefore these two materials could be managed together.

A company called Homasote produces multipurpose panels for building and sound proofing purposes. The panel is produced of 100 % post-consumer recycled newsprint, with paraffin wax working as the binder (Ecohaus Inc. 2009) According to Davis et al. (2003) there is a plant in Turkey manufacturing hardboard incorporating primary sludge to be used as core stock in furniture manufacturing.

Takáts and Takáts (2012) reported that fiber sludge from sulfate process is a good material as reinforcement for gypsum fiberboards. The process was characterised by a two-stage fluffing of the fiber sludge and addition of the lime sludge to reduce the setting time of the binder and to avoid formation of lumps and balling in the mixing process of the furnish. The new process and the product-fiberboard-is environmental friendly as material. The fabricated boards utilising natural gypsum, flue-gas desulfurization gypsum, phospho gypsum and stucco gypsum show comparable properties with those of published data of commercial gypsum fiberboards.

6.12.2 Moulded Pulp

Moulded pulp products have been produced since the early 1940s. Egg trays were the first product manufactured, but the range of products manufactured has increased since that time, to include fruit trays, simple containers, furniture corner protectors, industrial product packaging/partitions, and disposable products for catering and hospital applications. With the increasing use (and varieties of design) of moulded pulp products, production equipment and techniques have also evolved. New techniques for producing smooth surfaces, precise shapes and dimensions have been developed by the introduction of the Thermoforming process. Moulded pulp products have traditionally (and predominantly) been manufactured from recycled fibre, normally containing high percentages of pre-consumer newsprint (Kujala 2012). The pulp is formed on a mould, water partially removed by vacuum to form the shape after which the product is dried example in an oven. This application requires a maximum ash content of 10 % (Rothwell and Éclair-Heath 2007). West Fraser Timber's Alberta Newsprint Company supplied in 1997 seven percent of its sludge to manufacturing of molding egg cartons (Glenn 1997). However, no further information of this or more specific characterization of the sludge was found (Kujala 2012).

6.12.3 Millboard

Millboard is a recycled paperboard product. It is also manufactured from 100 % recycled fibre (Kujala 2012). It is a generic term referring to various high density and thickness board products. The product ranges in grammage from 1,000 up to 5,000 g/m^2, and is used in the automobile industry, shoe industry, furniture, luggage and leather products, and in the packaging and stationery industries. This type of use requires high fiber content and low ash content to guarantee wanted caliper (Rothwell and Éclair-Heath 2007). Millboard is a very hard board with high density and is manufactured by hard rolling (calendering) the semi-wet intermittent sheet on a solidboard machine. Successful use of the fibre fraction in Millboard would allow increased potential in the corrugated paper manufacturing industry to be a more viable option. The inclusion of high filler (ash) in the fibre fraction would however be detrimental to the final Millboard quality, particularly in terms of caliper (thickness). Trials to use sludge as an additive in millboard production were run with the fiber fraction of Aylesford paper mill sludge from recycled newsprint production. The incorporation of fiber fraction into millboard, in order to replace about 5 % of recycled fiber obtained from old corrugated containers, was successful. The final product was found to perform identically to the existing product and no influence on the final product properties were noticed (Rothwell and Éclair-Heath 2007).

6.12.4 Softboard

Softboard is primarily used for pin boards, and construction applications including insulation and fireproof applications (Kujala 2012). The raw material consists ideally of short fibers and has an ash content below 10 % for optimal thickness. Often softboard is manufactured from old newspapers. Softboard is produced from a thick wet pulp blanket which passes through a high intensity press and a drying tunnel (Rothwell and Éclair-Heath 2007). There is one manufacturer of softboard in the UK, the manufacture of which is a unique process, consisting of a wet-end forming area, where a large sheet of thick wet pulp is formed. From the wet-end the sheet passes to a high intensity press, (where water is removed and the calliper – thickness – is reduced to the required specification), and thereafter to the drying tunnel to reduce the water content to the final moisture level. Softboard is generally manufactured from old newspapers, which runs easily on the manufacturing machine, and which produce a final product with a high caliper. The raw material fibre is short in length and grey in colour, and must be as low in ash as possible (ideally less than 10 %) to obtain the maximum caliper in the final product. Test trials to use the Aylesford mill fiber fraction, in softboard production were conducted in the U.K. The final product was found to be weaker and denser than the existing product. The customer was willing to continue trials, if an ash content of below 10 % was achieved (Rothwell and Éclair-Heath 2007). According to Goroyias et al. (2004) softboard produced from paper waste streams containing around 80 % sludge and 10 % other fibres is possible. The other fibres can be either MDF fibre or virgin wood fibre. MDF fibre is preferred to achieve further cost savings. It is assumed that both primary and deinking sludge can be used for this application option due to their confirmed high amount of organic (fibres) content. Using sludge can replace virgin wood fibre. Virgin fibre costs are about £50–70/ton. The energy savings by using the rejects are around 0.10 GJ/ton$_{\text{wet}}$ and 10 kWh/ton$_{\text{wet}}$.

6.13 Mineral-Based Products

Reuse possibilities of deinking sludge and other recovered paper residuals depends on the composition of inorganic compounds. Common inorganics in deinking sludge are calcium carbonate and clay. In the combustion ash of deinking sludge, calcium oxide and sintered clay are primary components. Generally, three methods had been used, in 1994, to use (paper mill) sludge in building material industry. One of these was the sludge use as a feedstock to cement kiln. Another option was to use sludge in cementitious composites, where the use of organic fibers would increase the durability of the product and reduce cracking related to shrinking. In these studies it was concluded that combining Portland cement with deinking sludge could contribute to a composite material suitable for building blocks, wallboards, panels and such. Also use of sludge in the production of lightweight aggregate was studied (Weigand and Unwin 1994). In the reference document of Best available technology for pulp and paper industry (European commission 2001), it has been proposed to use sludge in cement industry, brick production, building industry, as well as for road construction. In addition to the options mentioned, following options seem to be possible: building board, glass or lightweight aggregate. Fire resistant products manufacturing with sludge is also possible. However, the selection of a real option is influenced by a number of factors (Tim et al. 2000; Weigand and Unwin 1994).

6.13.1 Cement and Cementitious Products

Cement is a universal term meaning binder, a material that sets independently binding other materials together. There are different kinds of cements, others that are called hydraulic because they harden as a result of hydration, an inorganic chemical reaction which provides example Portland cement its strength. Lime and gypsum plaster can be considered to be non-hydraulic cement, as they develop strength only in dry circumstances. The basic raw materials in the production of cement are limestone (CaCO3), clay, sand and iron ore. The proportion of limestone in kiln feed is about 80–90 % and clay 10–15 % (British geological survey 2005). According to Achternbosch et al. (2005) proportions of $CaCO_3$ and clay in raw meal are about 80 and 20 %. Also silica and aluminum can be beneficial in the process. When the raw materials are burned at a temperature of 1,400–1,500 °C, they form calcium, silicon, aluminum and iron oxides (Göttsching and Pakarinen 2000). The hard substance from the kiln is called clinker and is mixed with gypsum to produce Portland cement. The gypsum prevents cement from flash setting.

Usually, the proportions are 95 % of clinker to 5 % gypsum (British geological survey 2005). Residues from WWTP that are high in inorganics can contain significant quantities of these substances. Residues from recycled paper manufacturing consist mainly of inorganics, and the deinking sludge can provide components like silicon dioxide and aluminum which are beneficial for the process. Boiler ash from the wood and WWTP residues are found to be suitable for cement and brick manufacture, as long as the carbon content of the ash is below 6 %. In Portland cement production, the fly ash of newsprint mill is used. Ash can contribute to the process as a source of calcium, aluminum and silica (Bird and Talbert 2008). The main ingredients of cement production, CaCO3 and clay, are present in deinking sludge in high proportions, so theoretically the sludge could be beneficial material to the process. After incinerating paper sludge at approximately 800 °C, the resultant fly ash may contain reactive silica and alumina (in the form of metakaolin) as well as lime (CaO) which contributes chemically to the Portland cement ingredients. Paper sludge ash is therefore potentially suitable as an ingredient in:

- The cement kiln feed, contributing calcium, silica and alumina
- The manufacture of blended cements

According to Wiegand and Unwin (1994) in 1994 there was one mill practicing sludge reuse in cement production full-scale. They sent all their primary sludge and coal boiler ash to a cement manufacturer to fulfill 2 % of the kilns total feedstock. Champion International's Hamilton plant in Ohio, United States, used its sludge and boiler ash in 1997 as filler in cement manufacturing (Glenn 1997). The sludge Champion International provides to Portland cement manufacturing is primary sludge from a nonintegrated paper mill, dried with a rotary dryer to 85 % solids content (Hardesty and Beer 1993). Because the wastewaters and therefore sludge originates only from the papermaking process, it is expected to include high amount of inorganics and fibers in good condition. The inorganic content of the sludge was 40 % clay and 19 % limestone. However, the fiber proportion was 30 % compared to 7 % deinking sludge (Hardesty and Beer 1993). Most likely, there is not a lot of stickies and adhesives in the sludge, making it easier to dry and handle than sludge from a deinking process.

Ahmad et al. (2013) examined the possibility of using waste paper sludge ash as partial replacement of cement for new concrete. In this study waste paper sludge ash was partially replaced as 5, 10, 15 and 20 % in place of cement in concrete for M-25 mix and tested for its compressive strength, tensile strength, water absorption and dry density up to 28 days of age and compared with conventional concrete. From the results obtained, it is found that Waste Paper Sludge ash can be used as cement replacement up to 5 % by weight and particle size less than 90 μm to prevent decrease in workability. Further waste paper sludge has very high calorific value and could be used as a fuel before using its ash as partial cement replacement.

According to Göttsching and Pakarinen (2000), both deinking sludge and ash from combustion of deinking sludge can be used as secondary raw material to the kiln. Deinking sludge ash from fluidized bed combustion can also be used as a hydraulic additive to cement clinker (Göttsching and Pakarinen 2000). Champion International's Hamilton plant in Ohio, United States, used its sludge and boiler ash in 1997 as filler in cement manufacturing (Glenn 1997). The sludge Champion International provides to Portland cement manufacturing is primary sludge from a non integrated paper mill, dried with a rotary dryer to 85 % solids content (Hardesty and Beer 1993). Because the wastewaters and therefore sludge originates only from the papermaking process, it is expected to include high amount of inorganics and fibers in good condition. The inorganics content of the sludge was 40 % clay and 19 % limestone. However, the fiber proportion was 30 % compared to 7 % deinking sludge (Hardesty and Beer 1993). Most likely, there is not a lot of stickies and adhesives in the sludge, making it easier to dry and handle than sludge from a deinking process.

The use of deinking sludge in an application is dependent on how well the materials in sludge correspond to raw material proportions in the targeted application. As one universal recipe for cement production does not exist, the proportions of ingredients may vary. Typical chemical composition of cement is shown in Table 6.14 (Gineys et al. 2010). When comparing chemical compounds of deinking sludge ash to the chemical composition of cement, one can see that inorganic part of

Table 6.14 Chemical composition of cement

Compound	wt-%
CaO	68,07
SiO_2	22,98
Al_2O_3	4,73
Fe_2O_3	2,72
MgO	0,8
K_2O	0,69

Based on Gineys et al. (2010), Kujala (2012) sludge provides most of compounds present in cement. There are no guidelines or trace element threshold limits for heavy metals in cement or cementitious products. This is probably because trace elements are mostly bound to cement during the hydration and therefore their leachate is minimal and cement isn't considered to be hazardous to humans or to the environment. There was however a study in which the use of waste materials in cement production and their influences on trace elements in cement were examined (Achternbosch et al. 2005). These researchers concluded that trace element concentrations in cement would increase, but no maximum or threshold values for them were set. Maximum trace element concentrations in Portland cement are – Cadmium, 6 ppm; Copper, 98 ppm; Zinc, 679 ppm; Lead, 254 ppm; Nickel, 97 ppm; Chromium, 712 ppm (Achternbosch et al. 2005).

Sludge use in building products has followed three general techniques. One method is the use of sludge as a feedstock to a cement kiln. Raw materials used to produce cement can include calcium carbonate, clay, silica and smaller amounts of aluminium and iron. Some sludges contain significant quantities of these materials. Two companies have extensively investigated this alternative and one mill currently practices this on full scale (Bajpai et al. 1999). The mill sends all its primary sludge and all its coal boiler ash to the cement manufacturer. This is a combined total of approximately 100 tons/day. For the kiln involved, this amount of material represents only about 2 % of the total feed stock.

Addition of sludge into cement production process allows utilization of both energy and material content of sludge. Firstly, energy content of sludge is utilized in the process when sludge is calcined together with ordinary raw meal and, secondly, ash left becomes a part of cement clinker. DS is more appropriate for the process than WWTS due to its content of both organic and inorganic components (European Commission 2010). Use of sludge in cement production process has benefits and drawback. Primary desired chemical elements for cement production are lime – calcium oxide that comes from $CaCO_3$, silica – SiO_2, alumina – Al_2O_3, and iron. Thus, deinking sludge or its mixture with primary or secondary sludge can easily fulfill the need on those chemical elements listed in European Commission (2010). On the other hand, technological problems caused by sludge use could be, firstly, because of low heating value of sludge, if compared to heating value of conventional fuel used for cement production and, secondly, because of high MC.

Another alternative is the use of sludge in cementitious products. Lot of work has been done on the use of organic fibers including wood pulp in cementitious composites. The advantages include increased durability and pumpability as well as reduced shrinkage-related cracking (Thomas et al. 1987). Two studies undertaken to assess the performance characteristics of composites which included paper industry sludge concluded that a composite material potentially useful in building blocks, wallboards, panels, shingles, fire retardants and filler materials for fireproof doors could result from combining Portland cement with sludge from deinking mill. It was found that mixtures including Portland cement, ash, sand and sludge yielded a compressive strength comparable to conventional concrete and superior flexural strength (Thomas et al. 1987; Hall 2011a, b).

6.13.2 Cement Mortar Products

Cement mortar is based on organic filling material, produced from formation and solidification of a mixture consisting of Portland cement, DS, chemical additives, and water. For building blocks production from cement mortar, technical specification TU 69 USSR 82–84 "Building blocks made of cement mortar for farm building" was created in Ukraine. The material was classified as insulating concrete (Pavel 2008). Average consumption of raw materials for the production of 1 m^3 of building blocks is: Portland cement M400 – 230 kg, sludge (as absolutely dry solids) – 500 kg. Production of blocks is performed by blending of wet sludge with Portland cement without water. Compressing of the mixture produced is done at pressure of 0.08–0.12 MPa. Mode of solidification of blocks is natural or artificial drying. To prevent dried blocks from water absorption, its opened surface is recommended to be covered with repellent (Pavel 2008).

Pilot plant was established in Ukraine to produce such building blocks. To check real applicability of the blocks, two houses were built: one was built as a monolithic construction, and another one from small piece materials. Thermo technical properties of the houses were analyzed. Heat conductivity was equal to 0.2 W/(m·°C) for material density of 600–650 kg/m^3, and 0.3 W/(m·°C) for material density of 750–800 kg/m^3. Heat protection was higher than required by legislation. It enabled to decrease thickness of the walls made of the blocks until 18 cm (Pavel 2008). Application of sludge in cement mortar production was proven to be successful in terms of energy efficiency of the buildings made of the cement blocks.

Yan et al. (2011) have studied the use of sludge as an additive in cement mortar products and the effects on the physical and mechanical properties of the mortar. Cellulose is a known retarding concrete admixture, and in the study 20 weight-% of sludge in the mixture delayed final setting time. The compressive strength with the sludge was 62 % of that of the reference, at 20.5 MPa but was considered to be in range for masonry product usage. The sludge did not appear to make any difference in long-term cement hydration or hardened paste properties. The sludge proportion increased drying shrinkage and the volume of permeable voids in the final product. Dissolved lignin can work as an air entraining admixture, providing better workability and consistency and freezing durability to the concrete. A conclusion of the study was that up to 2.5 weight-% deinking sludge did not affect the physical or mechanical properties of the mortar significantly and incorporating deinking sludge can be a favorable supplementary addition to the product (Yan et al. 2011).

6.13.3 Pozzolanic Material

Pozzolanic material or simply pozzolan is defined as "a siliceous or siliceous and aluminous material, which in itself possesses little or no cementing property, but will in a finely divided form – and in the presence of moisture – chemically react with calcium hydroxide at ordinary temperatures to form compounds possessing cementitious properties" (Brunjes 2008; Deviatkin 2013; Gracia et al. 2008; Pozzolanic additives to cement 2010). Pozzolanic activity means to react with lime at normal conditions resulting in generation of viscous phases (MetaPro 2011). Moreover, addition of 15 % of pozzolan makes bonds with lime, thus, preventing bloom generation, that is known to be a problem in construction industry (Pozzolanic additives to cement 2010). Hundebol in his patent (1994) described a process of pozzolanic material production. In the ways given in the patent, kaolinic fraction of DS is transformed into a pozzolanic material in an environmentally safe and economic way as stated in the patent, pozzolan is a material consisting of silicon dioxide, aluminum oxide, iron oxide or similar to those mentioned. The most obvious method of pozzolan production is thermal treatment in a long rotary kiln as described in a patent (Hundebol 1997), however, malodorous gases can be generated, as well as external fuel will be required for the process. The scheme of pozzolan production is presented in Fig. 6.3 (Hundebol 1994). The use of fluidized bed boiler for pozzolan material production at the same temperatures as in the previous patent was proposed to activate pozzolanic properties of deinking sludge. Utilization of deinking sludge in the production of pozzolanic material had positive influence on mortars properties prepared with sludge addition (Frias et al. 2011).

The use of paper deinking sludge in pozzolanic material manufacture permits a disposable residue to be included in the cycle of the materials. A study on the reuse of paper de-inking sludge, undertaken in Spain, shows its potential as raw material for yielding a product with pozzolanic activity (Gracia et al. 2008). This study

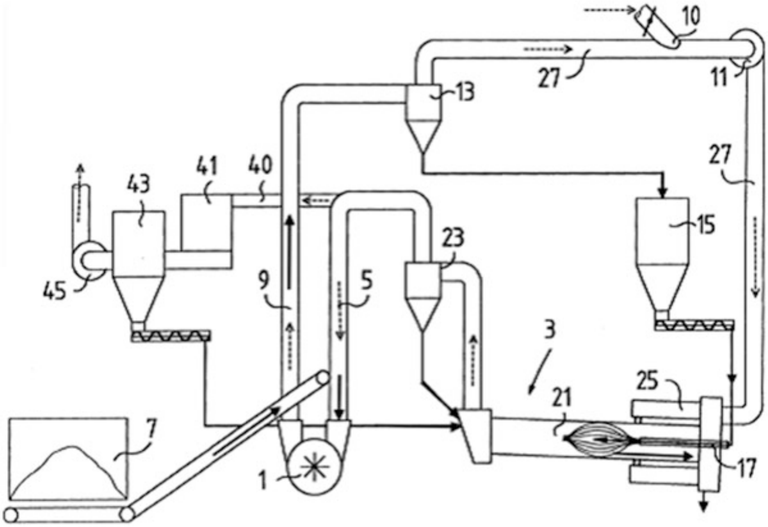

Fig. 6.3 Scheme of pozzolan production. Drier-miller (*1*); Kiln system (*3*); Duct (*5*); Duct (*9*) Separator (*13*); Air intake (*10*); Fan (*11*) Duct (*27*); Cooler (*25*); Storage silo (*15*); Burner (*17*) Rotary kiln (*21*); Cooler (*25*); Cyclone (*23*) Exhaust fumes (*40*); Economizer (*41*); Flue gas treatment system (*43*) (Based on Hundebol 1994; Deviatkin 2013)

established that an optimal condition for transforming paper de-inking sludge into a pozzolanic addition is achieved at 700 °C maintained for 2 h. Under these conditions, the organic matter disappears and the calcined sludge becomes active by transforming kaolinite into metakaolinite. The calcined product exhibits high pozzolanic activity. The researchers analysed mechanical, physical and chemical properties of a blended cement containing 90 % (in mass) standard Portland cement (CEM I-52,5N) and 10 % of the pozzolanic addition obtained from controlled calcination of paper de-inking sludge (for 2 h at 700 °C). When the blended cement is compared with a cement containing 100 % standard Portland cement, the following conclusions can be mentioned: a significant gain in compressive strength from 7 days on, a sooner initial setting time, as well as a reduction in SO_3 percentage. In any case, the new blended cement complies with requirements set out in Spanish/European standard UNE EN 197-1-Part 1.

Pera and Ambroise (1998) presented a research carried out to convert paper sludges from de-inking and water-treatment processing plants into a pozzolanic product usable in the cement and concrete industries. Paper sludges contain inorganic fillers like ground limestone, kaolinite, clay, and organics. The process consists in heat treatment in the range of 600–700 °C in order to transform the kaolinite present in the sludge into metakaolinite, a very reactive pozzolan, without the formation of large amounts of free lime (due to the decarbonation of the calcite also present in the sludge). In addition, the organic compounds in the sludge must be burned off and their presence reduces the use of fossil fuels to reach the temperature needed for processing. The results of both laboratory and field tests show the feasibility of the process. A reactive pozzolan is obtained when the amount of kaolinite present in the sludge is higher than 20 % of the dry inorganic phase. This metakaolin is sometimes more reactive than that obtained by calcining pure kaolinic clays, and can be used to enhance the durability of concrete in severe environments.

6.13.4 *Concrete*

Concrete is one of the most widely used construction materials in the world. It is a composite material consisting of aggregate, usually sand or gravel, mixed with water and cement. Cement works in the mixture as a binder. However, the production of portland cement, an essential constituent of concrete, leads to the release of significant amount of carbon dioxide, a greenhouse gas; 1 ton of portland cement clinker production is said to create approximately 1 ton of carbon dioxide and other greenhouse gases. Environmental issues are playing an important role in the sustainable development of the cement and concrete industry. For example, if we run out of limestone, as it is predicted to happen in some places, then we cannot produce portland cement; and, therefore, we cannot produce concrete and all the employment associated with the concrete industry goes out-of-business. Limestone powder is sometimes interground with clinker to produce cement, reducing the needs for clinker making and calcinations. This reduces energy use in the kiln and carbon dioxide emissions from calcinations. A sustainable concrete structure is one that is constructed so that the total environmental impact during its entire life cycle, including during its use, is minimum. Concrete is a sustainable material because it has a very low inherent energy requirement, is produced to order as needed with very little waste, is made from some of the most plentiful resources on earth, has very high thermal mass, can be made with recycled materials, and is completely recyclable. Sustainable design and construction of structures have a small impact on the environment. Use of "green" materials embodies low energy costs. Their use must have high durability and low maintenance leading to sustainable construction materials (Mymrin et al. 2009). High performance cements and concrete can reduce the amount of cementitious materials and total volume of concrete required. Concrete must keep evolving to satisfy the increasing demands of all its users. Reuse of post-consumer wastes and industrial byproducts in concrete is necessary to produce even "greener" concrete. Use of coal ash, rice-husk ash, wood ash, natural pozzolans, GGBFS, silica fume, and other similar pozzolanic materials can reduce the use of manufactured portland cement clinker; and, at the same time, produce concrete that is more durable. "Greener" concrete also improves air quality, minimizes solid wastes, and leads to sustainable cement and concrete industry.

Blawaik and Raut (2011) conducted a test with concrete and waste paper pulp and Yan et al. (2011) conducted study with mortar and sludge. These researchers observed that compressive, splitting tensile and flexural strength of concrete at 28 days increased up to a 10 % proportion of waste paper pulp. Further mixing of pulp into the concrete reduced all the strength properties gradually (Blawaik and Raut 2011). The increase in strengths could be explained by differences in the fiber proportion. Long, good condition fibers are expected to give better strength characteristics than short damaged fibers. Blawaik and Raut (2011) conducted research for 28 days, and Yan et al. (2011) determined the strength properties after 90 days.

Sludge use as an additive in cement and cementitious products has been studied and implemented to some extent. The results are promising. The primary sludge already has a use in the cement industry, as does the ash recovered from sludge incineration. The studies about deinking sludge in cement production are somewhat controversial, as some researchers conclude that deinking sludge hinders the development of mechanical strength in products and others that up to a 10 % sludge proportion sludge can be beneficial to products. All the studies however imply that the use of deinking sludge in cementitious products is preferable. None of the studies reported sludge fractionation to be obligatory, and that the sludge was used as such.

The use of paper-mill pulp in concrete formulations was investigated by Naik and Moriconi (2005) as an alternative to landfill disposal. The cement has been replaced by waste paper sludge accordingly in the range of 5–20 % by weight for M-20 and M-30 mix. By using adequate amount of the waste paper pulp and water, concrete mixtures were produced and compared in terms of slump and strength with the conventional concrete. The concrete specimens were tested in three series of test as compression test, splitting tensile test and flexural test. These tests were carried out to evaluate the mechanical properties for up to 28 days. As a result, the compressive, splitting tensile and flexural strength increased up to 10 % addition of waste paper pulp and further increase in waste paper pulp reduced the strengths gradually. The research on use of paper sludge can be further carried out in concrete manufacturing as a new recycled material.

Studies have shown that waste paper sludge ash (WPSA) contains considerable quantities of alumino-siliceous material and can be used as a unique cement replacement material. Sharipudin et al. (2012) attempted to incorporate incinerated WPSA and fine recycled concrete aggregate (FRCA) to produce new types of lightweight foamed concrete. They studied the effect of the inclusion of WPSA and FRCA replacement as cement and sand content respectively with respect to the compressive strength performance of foamed concrete. The study exhibited that WPSA and FRCA seems to contribute to a favorable compressive strength of foamed concrete.

Recently, Nazar et al. (2014) conducted study on the use of paper mill sludge as recycled materials and additives in concrete mixes for use in construction projects. The study had to provide the assurance that the concrete produced had the correct mechanical strength. Concrete mixes containing paper mill sludge were prepared, and their basic strength characteristics such as the compressive strength, flexural strength, ultra pulse velocity and dynamic modulus elasticity were tested. Four concrete mixes, i.e. a control mix, and a 10, 20, and 30 % mix of paper mill sludge as cement replacement for concrete were prepared with a DoE mix design by calculating the weight of cement, sand and aggregate. The performance of each concrete specimen was compared with the strength of the control mix. As a result, when the percentage of paper mill sludge in the concrete increased, the strength decreased. Overall, a high correlation was observed between density and strength of the concrete containing paper mill sludge.

6.13.5 Ceramic Material

Several papers have been published on the utilization of deinking sludge in the production of ceramic materials. The results vary greatly. The reason could be attributed to different production processes. Behavior of ceramic bricks made of clay and deinking sludge was examined by several researchers (Radovenchik et al. 2010; Demir et al. 2005, Sutcu and Akkurt 2009). In all cases the results were mainly negative. Radovenchik et al. (2010) used deinking sludge with moisture content of 68 %, ash content of 10 %. Amount of deinking sludge added varied from 0.5 to 6.0 % (on dry basis). Increased amount of deinking sludge incorporated resulted in increased moisture content of furnace feed by 3.0 % that resulted in additional fuel use. However, due to increased porosity of the material produced, less energy was required for its drying since atmospheric drying was more efficient. In addition, shrinkage rate that shows how much the material will be reduced in volume with time increases when more sludge is used. This fact causes problems since shape and dimensions of the material produced are not stable. Strength properties decreased dramatically. The reductions were 62 % when 5.5 % of sludge was added. The only positive result is decreased density of the material (Radovenchik et al. 2010). Demir et al. (2005) obtained similar results. In addition to parameters mentioned above, water absorption ability of the material produced was analyzed. It was found to increase by 37 % when 10 % of deinking sludge was added. Increased water absorption capacity can, improve setting of concrete with the bricks to make the construction stronger and increase weight of bricks.

Sutcu and Akkurt (2009) studied similar process with deinking sludge containing about 60 % of inorganic matter and moisture content of 65 %. It was found that due to increased porosity of the material produced with 30 % of sludge added, density of material and thermal conductivity decreased by 67 % and 50 % respectively. Moreover, mechanical properties of the material intended to be used in the construction industry decreased much. So, compressive strength of material where 30 % of clay was substituted with deinking sludge was about eight times lower in comparison to material made of ordinary ingredients (Sutcu and Akkurt 2009).

Asquini et al. (2008) studied production and characterization of sintered ceramics from paper mill sludge and glass cullet. Three different types of paper mill sludge were first incinerated and then attrition milled separately or mixed with glass cullet in varying proportions to obtain powders of different compositions. These powders were then dried, sieved, uniaxially pressed into samples and air sintered. Fired samples were characterized by density, water absorption, shrinkage on firing, strength, hardness and fracture toughness measurements; SEM and X-ray diffractions were also carried out to investigate microstructure and phase composition. Some sintered samples displayed fairly good physical and mechanical properties as a consequence of their low residual porosity and fine microstructure.

High quality ceramics can be produced as stated in the patent by Treschev et al. (2007). Following components and their volume-% were used:

- Weak clay – 64–66
- Deinking sludge – 6–9
- Ferrous waste – 7–13
- Keramzite foam clay – 15–20

The deinking sludge contained 60–80 % moisture and about 50 % ash content. Deinking sludge could be used for the ceramics production without deterioration of final product if produced as stated in the patent (Treschev et al. 2007). However, three other literature sources provided extensive information that incorporation of deinking sludge decreases properties of the final product.

Furlani et al. (2008) studied synthesis and characterization of ceramics from coal fly ash and incinerated paper mill sludge. Powders obtained from coal fly ash and paper mill sludge were milled alone or in mixture, pressed into specimens and sintered. Fired materials were characterized by density, water absorption, shrinkage on firing, crystal structure, microstructure, strength, hardness and toughness. Samples made with paper mill sludge alone had a high sintering temperature, contained extended fractures after sintering and were not further studied. All the other compositions, sintered between 1,130 and 1,190 °C, showed low water absorption, density below 2.6 g/cm^3, fair mechanical properties and contained several phases. More in particular, the material containing 25 wt.% of coal ash and 75 wt.% of powder from paper sludge displayed the best overall behaviour.

6.13.6 Lightweight Aggregates

Sludge has been also used in the production of lightweight aggregates (LWA) (Weigand and Unwin 1994; Meade et al. 1994). Aggregate is a term describing a collection of materials used as a filler in construction materials. Aggregates find use in cementitious products such as concrete, masonary, building blocks and asphalt. Sand and gravel or both are typical aggregate materials mixed with cement to produce concrete. Lightweight aggregate refers to a select group of materials which allow for reductions in final density while maintaining acceptable strength properties. Products which sometimes incorporate LWA include concrete block, architectural panels and decorative stone. As a strong and lower density material, lightweight aggregate is used in pre-cast concrete to reduce the load by 25–35 % in bridges and high-rise buildings. It is used in concrete blocks to reduce their weight and increase worker productivity. Concrete made with light weight aggregate also has higher strain capacity and lower shrinkage. Different ranges of regular and light weight aggregates are mixed depending on the specifications required. Used as a geotechnical fill it posses perfect characteristics to reduce dead loads and keep from soil thrusting while at the same time increasing drainage. It is inert and chemically stable so it can be used in landfills.

In the production of LWA, required amount of deinking sludge is taken from a silo and then is mixed together with cement in a mixer. Mixing is implemented in a thorough way. Well mixed blend is then sent to a pelletizing machine where granules are produced. Last stage of the whole process is drying of the material produced at the temperature of 100–105 °C (Liaw et al. 1998). Composition of the sludge used for lightweight aggregate production is presented in Table 6.15 (Liaw et al. 1998). Testing of the material produced included determination of volumetric specific gravity. The results from analyses showed that the best mixing time is about

Table 6.15 Composition of the sludge used for lightweight aggregate production

Moisture content of sludge, %	75.4
Loss of ignition of dried sludge, %	70.11
Ash Content, %	30.0
SiO_2	37.99
Al_2O_3	51.72
Fe_2O_3	3.04
CaO	5.09
MgO	3.10

Based on Liaw et al. (1998)

10 min when any cement/deinking sludge ratio can be used for material production to obtain lightweight aggregate with volumetric density less than 1.0. If material has been mixed for 40 min then only cement/ deinking sludge ratio less than 0.6 can be used. Thus, the longer raw meal is mixed, the more density of the material produced (Liaw et al. 1998). Test bricks with dimensions of 40 × 10 × 10 cm were prepared using aggregate material produced from sludge. One brick weighted 5.4 kg that is 23 % less compared to the one produced from concrete (7 kg). Compressive strength can reach 1,780 psi, that is almost the same as set for load bearing concrete masonry units in the Standard Specification for Concrete Brick (Hanson 2009). To recapitulate, porosity of light-weight aggregate produced is lower, if mixing time is shorter. Moreover, incorporation of sludge has positive influence on light-weight aggregate produced because of decreased density, while compressive strength stays in acceptable limits. An alternative to disposing of paper mill waste in landfills has been developed by the Greengrove Corporation (Meade et al. 1994). The process consumes 686 tons of paper mill sludge and more than 124 tons of ash to produce 250 tons of lightweight aggregate, but can be adapted to other volumes of waste.

6.13.7 Plasterboard

Paper sludge could also be utilized as an additive in the plasterboard. Plasterboard is made from gypsum which is a naturally occurring rock. “Stucco” is made by crushing gypsum, which is heated to remove the water of crystallization reducing it to ½ H_2O. In this form it becomes a free flowing powder called stucco. Plasterboard is made by pouring stucco (mixed with a stoichiometric quantity of water) onto a moving sheet of paper, and gluing a second sheet to the upturned paper edge. The paper thus forms a sandwich, and the plaster is rolled flat and allowed to set (crystallize) while moving down a long conveyor. The plasterboard is finally dried with hot air after the crystallization reaction has ended. Initial flow is important to spread the plaster. Setting time is critical (the plaster must be set at the end of the line) and there is a cost to drying if more water than the ideal is used in the process. The rate at which the plasterboard sets can be increased by additives. Adding gypsum to the stucco increases the rate of setting at the lowest cost. In theory, the fibrous material in sludge could improve some strength properties or provide a porous structure to the final product and therefore lower density. According to Hall (2011a), paper pulp is currently added to plaster to improve the tensile strength of the core, yet the composition and type of sludge is not specified. 50 % of gypsum panel is air, to minimize the product weight and to make it easier to work with. The porous structure is produced by foaming agents. Vermiculite or clay may also be added to the product to increase enhance fire resistance (Hall 2011a). The fiber proportion could be useful at strength improvement but is possibly harmful for the fire-resistance of the plaster. When produced at high enough temperatures, the organic matter burns leaving pores in the structure. However, most likely the drying temperature of 250 °C is not high enough. Clay in the sludge could also be used in board production, but should be as pure as possible from contaminants. Hall (2011a) states that each additive of plaster can be a maximum of 0.5 % gypsum powder, so the plasterboard production can even in theory, provide reuse for only a limited amount of sludge.

Another gypsum product to make with waste paper is gypsum fiberboard, in which there are no conventional board sheets around the gypsum board, but 18 % of the gypsum material is replaced by ground recycled fiber to provide reinforcement (Venta 1997). Recycled fiber reinforcement of gypsum composites have also been studied by Carvalho et al. (2007) but in this study the cellulose fiber content came from repulped kraft paper cement bags which has quite different composition to deinking sludge. The study can only provide indicative information on mechanical properties of fiber cellulose composite.

A patent filed in United States in 1996 described the drywall and building block manufacturing process with deinking by-product reinforcement. This method involved addition of deinking by-products at a consistency of 3–10 weight-% to stucco. The process achieved a deinking-based fiber loading in drywall up to 30 weight-%, and provided better bending strength property to the final product. The composition of the by-products was not disclosed in the patent (Tran 1993).

Rothwell and Éclair-Heath (2007) conducted trials with the filler fraction of Aylesford mill sludge to be used in acoustic plasterboard production. The filler content of stucco-filler mixture was at first 10 %, but later the filler content of 5–7 % was found to be optimal. Also in these trials it was noted that extra material worked as retardant, the setting time of mixed plasterboard being 11 min compared to normal time of about 8 min. The results showed that incorporation of filler in plasterboard was successful. It was speculated whether a purer filler fraction would give even better results (Rothwell and Éclair-Heath 2007).

6.13.8 Insulating Material

Utilization of deinking sludge in the production of insulating material has been described by several researchers (Cavaleri and Contu 2011; Morozova and Shargatov 2001; Zvyagina and Pushnoy 1996; Neckermann and Wooding 1982). Deinking sludge can be widely used in the production of thermal and acoustic insulating material. One of the promising innovations is the use of paper mill sludge as heat insulation material with a thermal conductivity factor lower than 0.055 W/m^{2}K. This is comparable to insulation materials available on the market currently. Using the paper mill sludge for production of construction and insulation boards has been topic of The Waste and Resources Action Programme (Goroyias et al. 2004).

Different production processes are known with different additives. Thermal conductivity coefficient for obtained materials was between 0.07 and 0.1 Wt//(m·oC) that is somewhat similar or less than that of lightweight concrete

(0.1–0.3 Wt//(m·oC) (The engineering toolbox 2013). Cavaleri and Contu (2011) describe production of thermal and/or acoustic insulation material in the form of flakes. It was stated that sludge containing long fibers is more appropriate for the production process. Presence of inorganic content decreases insulating properties of the material being produced. Deinking sludge with moisture content between 25 and 60 % is used. The sludge is stored in a silo. The silo is used as a mixing point where liquid additives, such as fungicides, flame retardants, and coloring agents are added to sludge, if required. Then the mixture is sent for drying process to a drying machine and after that the mixture is hammered in a crusher until its shape is flake-like. Also, powder additives can be added in the crusher, if required (Cavaleri and Contu 2011). Another way to produce insulating material from deinking sludge is to produce it in the form of boards (Morozova and Shargatov 2001). The material can be used for manufacturing of wall panels used for thermal insulation of residential, industrial, and other buildings, as well as for thermal insulation of hot and cold water and steam pipelines. Another way of insulating material production has been described by Zvyagina and Pushnoy (1996). For the production of insulating material following components and their amounts in weight-% are required: cement – 22–30, granulated deinking sludge with density of 300–400 kg/m^3 – 56–66, and latex – 12–14.

6.14 Wood Adhesive

The use of the paper mill sludge as a paper and wood adhesive is an interesting concept (Likon and Trebše 2012; Geng et al. 2007b). Three major types of paper mill sludge, primary sludge, secondary sludge and deinking paper sludge were characterized and evaluated as adhesive fillers. Plywood panels were made of formulations with phenol formaldehyde and sludges. Panels with phenol formaldehyde/primary sludge and phenol formaldehyde/secondary sludge formulations had higher dry and wet shear strengths than those made with phenol formaldehyde/Cocob® formulation. All wood failure values were comparable. Dry and wet shear strengths of the panels with phenol formaldehyde/deinking paper sludge formulation were comparable to those of the phenol formaldehyde/Cocob® panels (with Cocob® as a commercial filler), but the former displayed a much lower wood failure value. Owing to this fact and its high ash content, deinking paper sludge was not evaluated further as a potential component of adhesive formulations. Compared with secondary sludge, primary sludge resulted in higher dry and wet shear strengths and higher wood failure values. However, granular secondary sludge was easier to disperse into the resin component than fibrous primary sludge, and the phenol formaldehyde/secondary sludge formulation was more easily dispensed on aspen veneer sheets than the phenol formaldehyde/primary sludge formulation. Secondary sludge alone displayed adhesive properties with 0.87 MPa of dry shear strength, but primary sludge alone did not exhibit any bond strength. Primary sludge and secondary sludge were further evaluated for their general thermal behavior and major functional groups using differential scanning calorimetry and Fourier transform infrared spectrometry, respectively.

Pervaiz (2012) explored the bio-adhesion characteristics of secondary paper sludge and its value-added utilization as wood adhesive. Several analytical studies on the secondary sludge from municipal waste treatment plants have confirmed the presence of biopolymers, especially protein, in significant quantities and suggested its use as poultry feed (Hwang et al. 2008; Jung et al. 2001, 2002; Lau 1981; Lerch 1991; Lerch et al. 1993a, b). Paper mill secondary sludge, accounting for almost one half of total mixed sludge, is believed to be similar to municipal effluent activated sludge in terms of microbial chemistry and contains 30–40 % proteins on dry weight basis (Edalat Manesh 2012; Jung et al. 2002). Although microbial and cellular studies have suggested use of secondary sludge as potential surface active agents and adhesives (Garcia-Becerra et al. 2010; Edalatmanesh et al. 2012; Edalat Manesh 2012) but current knowledge is devoid of any reference pertaining to protein recovery from paper sludge and its characterization except the work by Pervaiz (2012) which is the first of its kind, related to protein recovery from paper secondary sludge through a modified protocol in substantial quantities and its comprehensive characterization. After identifying extracellular polymeric substances as adhesion precursors through analytical techniques, Pervaiz (2012) carried out study to optimize protein recovery from SS and its comprehensive characterization. A modified physicochemical protocol was developed to recover protein from secondary sludge in substantial quantities. The combined effect of French press and sonication techniques followed by alkali treatment resulted in significant improvement of 44 % in the yield of solubilized protein compared to chemical methods. The characterization studies confirmed the presence of common amino acids in recovered sludge protein in significant quantities and heavy metal concentration was reduced after recovery process. The sodium dodecyl sulfate polyacrylamide gel electrophoresis analysis showed the presence of both low and high molecular weight protein fractions in recovered sludge protein. After establishing the proof-of-concept in the use of recovered sludge protein as wood adhesive, the bonding mechanism of protein adhesives with cellulose substrate was further elucidated in a complementary protein-modification study involving soy protein isolate and its glycinin fractions (Pervaiz 2012). The results of this study validated the prevailing bonding theories by proving that surface wetting, protein structure, and type of wood play important role in determining final adhesive strength. Recovered sludge protein was also investigated for its compatibility to formulate hybrid adhesive blends with formaldehyde and bio-based polymers. Apart from chemical cross-linking, the synergy of adhesive blends was evaluated through classical rule-of-mixture. The findings of this study warrants further investigation concerning other potential uses of recovered sludge protein, particularly as food supplements and also the economic implications.

6.15 Sorbent Production

WWTP residuals have been used as raw material in industrial absorbents which are available in market (Bird and Talbert 2008). For example, in Washington, United States, a company called International Absorbent Inc. has used paper mill sludges in oil absorbent materials since the 1990s (Glenn 1997). In 1996, a Wisconsin company called Thermo Fibergen had developed a system to recover long fibers and extract water from the sludge to use the fibers in papermaking and after that the material of short fibers and fillers were to be used as oil and grease absorbents, cat litter and granules. They had bought a company called GranTek which already had a continuous operation to take sludge at a consistency of 40 % solids, dry it and form granules to be further dried and screened (Glenn 1997). As sludge from a recycled paper process already includes relatively few long fibers, it might be a suitable raw material for these type of products, if it could be easily dried. Complete Spill Solutions also use WWTP residuals in their absorbent products.

Promising research has been conducted to use paper mill sludge as an oil sorbent material and use of the paper mill sludge as sorbent material is well documented, but currently the market was non receptive to such sorbent material due to cheap and efficient synthetic absorption material. The results of research studies have shown that paper mill sludge can be indirectly used as an active absorbent by converting it into activated carbon (Ben-Reuven 1997). It can be used as binding material for the removal of heavy metals ions from water (Battaglia et al. 2003; Calace et al. 2003; Hea et al. 2010; Ahmaruzzaman 2011), removal of phenols (Calace et al. 2002) and as an absorbent for hard surfaces cleaning (Lowe et al. 1988; Eifling and Ebbers 2006). A variety of the processes and different absorbent products have been developed for commercial purposes.

One of the processes which have been developed for the production of a floor absorbent, in the form of a granular product is known as the KAOFIN process and is patented (U.S. Patent 4343751) (Naresh 1980). In U.S. Patent 4374794, the sludge is evaporated, into pellets and dried at temperatures ranging from 100 to 150 °C, in order to form an oil absorbent material (Kok 1983). However, modern industry faces frequent and serious oil spills and subsequent sanitation demands high costs for sorbent materials. Offering a cheap and efficient natural material such paper mill sludge could become a welcome solution. The CAPS (Conversion of paper mill sludge into absorbent) is an eco-innovation solution in the 'market uptake' phase (Likon and Trebše 2012; Likon et al. 2011). This process uses the excess of the thermal energy which paper mills usually waste into the environment for sorbent production. In addition, CAPS uses paper mill waste as a secondary raw material and converts it into a high added value absorbent. The technology is relatively cheap, simple and easily replicable particularly in markets with a developed paper industry. It is based on drying of paper mill sludge to the point where it can be efficiently mechanically and/or chemically treated to release cellulosic fibers from its inorganic matrix. The humidity of the deinking and primary paper mill sludge lies between 50 and 70 %, whereas the content of cellulosic fibers is approximately 52 %

and the remaining portion is inorganic. After drying between 70 and 80 % of the solid content, paper mill sludge proceeds through special mechanical treatment. This stage is critical for the entire process due to the reason that in this section cellulose fibers are released from the inorganic matrix, which in turn allows material to float on the fluid surface. However, the mechanical treatment expanded the surface area, but the sorbency was not linear with regards to the surface area. The mechanical treatment was clearly connected to the breaking of the inorganic matrix into the dust. The dust dropped off from the absorbent, which led to the shrinking of the surface area but made the remainder of the surface more amenable to the substrate. More violent mechanical treatment during the production (fluffing) of the absorbent led the isothermal sorption of the absorbent to become similar to the isothermal sorption of the paper standard. The results showed that the appropriate treatment process particularly the mechanical treatment phase was very important for the conversion of the paper mill sludge into a sorbent. The life cycle's assessment study shows a reduction of carbon footprint for more than 14 times and reduction of water consumption for 372 kg, based on the production of sorbent material for cleaning 1,000 kg of oil spill. A conversion of the paper mill sludge into sorbent material prolongs paper products' life cycle for additional two cycles. A controlled incineration converts the used sorbent into inert meta-kaolin product which can be further used as hydrophilic sorbent material. In this way the papermill sludge's life cycle is efficiently closed.

Adsorption capacity and bioactivity of a novel mesoporous activated carbon (IIT Carbon) and bioactive (BAC_{IIT}) catalyst produced from paper mill sludge were evaluated by Khalili et al. (2002). Conversion of paper mill sludge to useful activated carbons and biocatalysts is a significant process since it reduces environmental problems associated with disposal of waste sludge, enhances wastewater treatment using carbons produced from industrial waste itself, and promotes conservation of the naturally available primary resources currently used to make activated carbons. Analysis was conducted using synthetic wastewater containing phenol and a commercially available activated carbon, sorbonorite 4 (used as reference carbon). Phenol removal was accomplished in batch and fluidized bed reactors containing mesoporous activated carbon, sorbonorite 4, and the produced bioactive catalysts. Isotherm adsorption data indicated that mesoporous activated carbon has a higher adsorption capacity and molecular surface coverage than sorbonorite 4 for phenol concentrations less than 10 mg/L. The mass transfer limitation was accounted for the lower adsorption capacity of the microporous carbon (sorbonorite 4) in dilute solutions. The fluidized bed reactor study, however, indicated similar but slightly lower phenol removal capability for the produced mesoporous carbon. While phenol removal efficiency of the carbons studied was in the range 65–70 %, the produced bioactive catalysts were able to remove up to 97 % of phenol during first few hours of operation. These results suggest that mesoporous carbon will feasibly be a good substitute for other commercially available activated carbons produced from natural resources, not only in physical adsorption processes, but also in fluidized bed bioreactors (FBB), used in biodegradation processes.

6.16 Filler in Nylon Biocomposite Production

Secondary sludge from pulp and paper mills can be considered as potential filler for composite industry (Fowler et al. 2006; Mohanty et al. 2002). Edalat Manesh (2012) studied the use of pulp and paper mill sludge as renewable and cost-cutting filler in the composite. The results of mechanical strength tests showed that a 10 % sludge content does not lead to any significant deterioration of either tensile or flexural strengths. Therefore, it was concluded that the secondary sludge may be used as filler to reduce the cost while maintaining the mechanical properties of Nylon.

Edalatmanesh et al. (2012) also investigated enzymatic modifications of the waste secondary sludge from pulp and paper mills to reduce the hydrophobicity and increase the molecular weight by lipase and laccase, respectively. The enzymatic modification was performed to enhance the reinforcing capability of the secondary sludge for further composite production. The lipid content of the secondary sludge, which was measured to be 6 ± 0.5 %, was hydrolyzed by lipase from Candida rugosa and the structural changes were followed by Fourier transform infrared (FTIR) spectroscopy. Laccase from Trametes versicolor was tested for its activity and reaction rate in the secondary sludge and the alkali-extracted lignin. Characterization of the sludge before and after the laccase treatment was carried out by FTIR spectroscopy. High pressure size exclusion chromatography (HPSEC) was applied to determine the molecular weight distribution of the lignin samples and also as a means for comparing modified and unmodified samples. Biokinetic parameters for the Michaelis-Menten kinetic model as a function of dissolved oxygen concentrations were determined The Km values were found to be 3.491 and 2.318 g/m^3 for sludge and the alkali extract, respectively. The FTIR results on the laccase-treated secondary sludge showed clear changes in the molecular structure, which was mainly attributed to the crosslinking reactions and generation of new bonds. Moreover, the HPSEC results demonstrated that laccase modifies the sludge by increasing the molecular weight. The manufactured nylon/sludge composites showed lower tensile strength for the lipase treated sludge/nylon composite. However, the laccase-treated sludge/nylon composite showed a statistically significant increase in the mechanical strength, which is attributed to the increase in the components molecular weight.

6.17 Landfill Cover Barrier

Significant progress has been made in the use of paper mill sludge as a material for land fill cover by replacing the clays or geo-composites (Van Ham et al. 2009). Several countries within the European Union (Finland, Sweden, etc.), as well as Canada, the United Stated and South Africa are investigating the possibility of implementing paper sludge as the hydraulic barrier material in landfill cover systems (Kortnik et al. 2008). Paper mill sludge behaves similar to a highly organic soil and has good chemical, hydrodynamic and geotechnical properties which make it an efficient impermeable hydrodynamic barrier for the land field cover (Zule et al. 2007). In North America, pulp and paper sludge has been used as a barrier layer in landfill final cover systems since 1975 (Moo-Young and Zimmie 1997; Moo-Young 1998). NCASI reports that between 1990 and May 2003 more than 29 landfills were closed using paper pulp sludge as the hydraulic barrier layer (Van Maltby 2005). These landfills ranged in size and composition from a 1.6 acre municipal landfill to a 30 acre industrial landfill. The NCASI report goes on to give a synopsis of the case histories of five of these landfills (Van Maltby 2005). The use of paper pulp sludge as part of the liner in the landfill expansion will reduce the need to truck in clay from a location offsite. In light of rising transportation cost this could prove to be a substantial monetary savings. The use of this pulp sludge will also continue to divert material destine to be place in the landfill to part of the landfill structure. Results have shown that the sludge barriers perform as well or better than the clay barriers (Weigand and Unwin 1994). Experience with the use of paper industry sludge as daily, interim and final cover for paper industry and municipal landfills is available. Worth mentioning is the experience of one recovered fiber processing mill. To demonstrate the utility of paper mill sludge as landfill-capping material, this mill constructed six test cells to compare the performance of primary sludge combined sludge and clay as hydraulic barriers (Weigand and Unwin 1994). Data from these test cells sufficiently supported a petition to the Massachusetts Department of Environmental Protection for a full-scale demonstration project. The project involved capping a 2 ha municipal landfill with combined mill sludge. To date, monitoring of cap performance indicates that the demonstration has been successful. Combining the paper mill sludge as hydrodynamic impermeable barrier with metal scoria as an oxidizing layer an efficient landfill capping system can be build. More than 21 different studies have been carried out in 2009 on the use of paper mill sludge in the landfill body (Rokainen et al. 2009). Moo-Young and Zimmie (1996) investigated the geotechnical properties of seven paper mill sludges for use as the impermeable barrier in landfill covers. Paper mill sludges have a high water content and a high degree of compressibility and behave like a highly organic soil. Consolidation tests reveal a large reduction in void ratio and high strain values that result from the high compressibility. Laboratory permeability tests were conducted on in situ samples, and these samples met the regulatory requirement for the permeability of a landfill cover. To determine the effectiveness of paper sludge as an impermeable barrier layer, test pads were constructed to simulate a typical landfill cover with paper sludge and clay as the impermeable barrier and were monitored for infiltration rates for 5 years. Long-term permeability values estimated from the leachate generation rates of the test indicate that paper sludge provides an acceptable hydraulic barrier. The characteristics of paper mill sludge have been exploited also for road bed construction for light loaded roads and tennis courts (Moo-Young and Zimmie 1996).

Howe Sound Pulp and Paper Limited Partnership (HSPP) was interested in beneficially using pulp and paper residuals generated by their facility in Port Melon, British Columbia in landfill closure. To facilitate use of pulp and sludge in landfill closure, SYLVIS (SYLVIS is a leading Canadian residuals management consulting and contracting firm with the sole focus of researching, recommending and implementing beneficial residuals management) worked with HSPP to complete a detailed assessment of the use of paper mill sludge as a barrier layer in the final landfill cover system. SYLVIS staff completed a site assessment and collected samples to assess the characteristics and quality of the pulp and paper sludge including hydraulic conductivity, organic matter, moisture, physiochemical properties and shear strength. Using computer simulation SYLVIS assessed the performance of final cover systems with different barrier layers: clay, sludge and a geomembrane over a clay layer. Based on the results SYLVIS concluded that both systems would make effective barrier layers in the final cover system. SYLVIS designed a fabricated soil layer for use in closure composed of residuals generated by the mill: wood, sludge and ash. Landfill closure using residuals generated on site provided significant cost savings for Howe Sound Pulp and Paper.

Kortnik et al. (2008) performed a laboratory test to evaluate the suitability of Slovenian waste paper sludge as landfill covering or sealing material. The sludges obtained from Slovene paper mills were examined for their geomechanical, chemical and sealing properties, as well as for their microbiological stability. The results of preliminary geotechnical studies of paper sludges indicate the expected relatively low strength properties ($14\ \text{kPa} < \sigma c < 81\ \text{kPa}$), high compressibility and a low coefficient of permeability $5.0*10-10 < k < 7.8*10-11$. The pH values of the eluates were within the permissible limits 6.6–7.0. The strength properties of paper sludges and various composites of paper sludge can be improved by additives (fly ash, bentonite, cement) and various composites of different types of paper sludges. The strength properties ($273\ \text{kPa} < \sigma c < 1{,}064\ \text{kPa}$) of these paper sludges were improved, but the coefficient of permeability k also increased slightly ($1.7*10-9 < k < 3.6*10-9$). These results complied with the prescribed conditions for mineral hydraulic protection layers used to cover the body of waste landfills and for sealing off the bottom of waste landfills for hazardous or non-hazardous waste.

Abitibi-Bowater Inc. (formerly Abitibi-Consolidated Company of Canada), with the guidance of SYLVIS and the authorization of the British Columbia Ministry of Environment, closed a 1.4 ha on-site ash landfill using a compacted sludge barrier system. The compacted sludge barrier layer appears to be performing in a manner equal to that of the conventional barrier systems (Van Ham et al. 2009). The use of compacted sludge in landfill closure presents a new opportunity for mills to use pulp and paper residuals in a low-cost, beneficial-use option.

6.18 Bacterial Cellulose and Enzymes

Fiber sludge consists mainly of cellulose and hemicellulose, and usually has a low content of lignin (≤ 5 %). Due to their composition and structure, fiber sludges are usually easy to be hydrolyzed enzymatically without prior thermochemical pretreatment, and could potentially yield hydrolysates with high glucose concentrations and low content of inhibitory compounds. A low content of inhibitory compounds should be advantageous for the bacterial strains used for production of bacterial cellulose. There are, however, drawbacks associated with enzymatic hydrolysis, especially the high cost for the hydrolytic enzymes used in the process. Cavka et al. (2013) investigated the appropriateness of waste fiber sludge for production of bacterial cellulose, and the possibility to combine the production of bacterial cellulose with production of hydrolytic enzymes useful for degradation of lignocellulose. The fiber sludges used in this study were originated from a pulp mill using a sulfate-based process (kraft pulping) and from a lignocellulosic biorefinery using a sulfite-based process. In addition, they investigated the metabolic preferences of the bacterium, Gluconacetobacter xylinus, used for the production of bacterial cellulose and the filamentous fungus, Trichoderma reesei (Hypocrea jecorina), used for enzyme production by analyzing the consumption of different components in the culture medium. The highest volumetric yields of bacterial cellulose from sulfate and sulfite process were 11 and 10 g/L (DW), respectively. The bacterial cellulose yield on initial sugar in hydrolysate-based medium reached 0.3 g/g after 7 days of cultivation. The tensile strength of wet bacterial cellulose from hydrolysate medium was about 0.04 MPa compared to about 0.03 MPa for bacterial cellulose from a glucose-based reference medium, while the crystallinity was slightly lower for bacterial cellulose from hydrolysate cultures. The spent hydrolysates were used for production of cellulase with T. reesei. The cellulase activity (carboxymethyl cellulase activity) in spent sulfate and sulfite hydrolysates reached 5.2 U/mL (87 nkat/mL), which was similar to the activity level obtained in a reference medium containing equal amounts of reducing sugar. It was shown that waste fiber sludge is a suitable raw material for production of bacterial cellulose and enzymes through sequential fermentation. The concept studied offers efficient utilization of the various components in fiber sludge hydrolysates and affords a possibility to combine production of two high value-added products using residual streams from pulp mills and biorefineries. Cellulase produced in this manner could tentatively be used to hydrolyze fresh fiber sludge to obtain medium suitable for production of bacterial cellulose in the same biorefinery. Sequential production of bacterial cellulose and enzyme would potentially give two high value-added products from a residual stream of very low or no value.

6.19 Cellulose-Based Specialty Products

The cellulose fibres recovered from recycling mill sludge may be applied to derive cellulose-based specialty products that can be used in various industries. The reaction route of including the extraction, intermediate formation and derivatization of cellulose is dependent upon a number of major factors. Hence, finding the optimum conditions for the process is a key aspect of the respective proposition. Cellulose fibres present in the recycling mill sludge amount for 50–55 % of the sludge content (Klyosov 2007). Thus, sludge may be treated as a source of cellulose, where the recovered cellulose fibres can be further used to produce derivatives and/or value-added products (Sultana 2013). Cellulose fibres can be recovered by oxidizing with sodium metaperiodate. The oxidation process not only extracts the cellulose fibres, but also activates the unreactive fibres for further derivatization by forming a biodegradable intermediate called dialdehyde cellulose (DAC). The DAC formed, which is highly reactive, can be further treated through crosslinking with substituent groups to form specialty cellulose-based products.

6.20 Nanocomposites

Cellulose nanocrystals have been evaluated as reinforcement material in polymeric matrices due to their potential to improve the mechanical, optical, and dielectric properties of these matrixes (Leão et al. 2007, 2012). Production of nanocellulose from paper mill sludge seems to be a great opportunity for the production of nanocellulose, because this sludge is already partially bleached, therefore it is interesting to use it as raw material due to its feature, less lignin, hemicellulose and other low molecular weight components, in a way to isolate the pure cellulose Leão et al. (2012) reported the successful isolation of nanofibrils from primary sludge by steam coupled acid treatment. The morphology of the sludge fibers from micro to nano scale were investigated by TEM proves for the successful separation of the well individualized sludge nanofibrils having the diameter less than 100 nm. The AFM analysis also proves the extraction of continuous nanofibers with a uniform diameter of approximately 12 nm, forming an extremely fine network. The TEM micrograph of the nanocomposite shows the sludge nanofibrils with the diameter below 20 nm were observed to be completely dispersed in the polyurethane matrix, preserving the nanofibril structure after composite fabrication. The XRD analysis confirms that cellulose nanofibril in the prepared nanocomposites preserves the original crystalline structure of cellulose (cellulose I). The mechanical properties of the developed sludge nanocellulose-polyurethane nanocomposite increase directly with the increase in nanocellulose content. The addition of only 4 wt.% cellulose nanocrystals, obtained polyurethane nanocomposite with tensile strength of 45.6 ± 1.4 MPa and modulus of 152.63 ± 1.3. The developed nanocellulose and its composites confirmed to be a very versatile material having the wide range of biomedical applications and biotechnological applications.

6.21 Paving and Fibrous Road Surfacing Additives

Paper mill sludge, is a good material in asphalt road pavement work (Kujala 2012). The fibers from paper mill sludge can withstand harsh processing, including high temperature for mixing with asphalt, as shown by the presence of fibers that were recovered after extraction (reported the Los Baños-based Department of Science and Technology Forest Products R and D Institute (DOST-FPRDI). Based on a research FPRDI conducted, it was concluded that the use of paper mill sludge, instead of imported paper additive in asphalt mix for road pavement, promises big savings. About P240,000 can be saved in paving a 1-km, 3-m wide, and 5-cm thick road using paper mill sludge instead of imported fiber. In the study, FPRDI assessed the possibility of using paper mill sludge and wastepaper as fiber additive in stone mastic (protective coating or cement) asphalt mix for road pavement. The sludge was dried, ground, and then sieved. The resultant material was mixed with aggregates and asphalt and then applied with heat, mixing, and mechanical compaction. The process turned out encouraging results. However, FPRDI conceded, "there is still a need for pilot-scale study to establish the economic viability of this technology as well as the long-term performance of the asphalt mix in road pavement."

Recycled fiber can be used in bitumen modifiers to achieve better properties for road surfacing materials. The fiber additive is known to reduce noise, and provide enhanced binding, elasticity and durability to the pavements. Rothwell and Éclair-Heath (2007) provided the fiber fraction of their research to a fibrous road surfacing manufacturer. The customer ran laboratory trials and found that the product failed the bitumen drainage test, and that for their products, a material with a moisture content below 7 % with an ash content below 20 % was required.

Mari et al. (2009) evaluated the properties of stone mastic asphalt mixtures made with paper mill sludge from four paper mills, as well as wastepaper, as fiber additive. Marshall specimens were prepared with asphalt content of 4.5, 5.0, 5.5, 6.0 and 6.5 % and sludge or fiber contents of 0.2, 0.3, 0.4 and 0.5 %. Properties tested were bulk specific gravity, stability, flow, air voids, voids in mineral aggregates and voids filled with asphalt. Effects of asphalt and fiber contents on flow and stability were analyzed statistically. Asphalt contents between 5 and 6 % and sludge or fiber contents between 0.3 and 0.5 % from any of the four paper mills resulted in Marshall specimens with properties generally passing the Department of Public Works and Highways specifications for both medium and heavy traffic road pavement.

The additive use of sludge can be considered as one of the applications that should be tested differently for the specific sludge type available. In these applications, the drying properties of sludge will most likely be the limiting factor for sludge reuse.

6.22 Other Options for Utilization of Waste

Paper mill sludge has been also used as growing media for soilless production. Traditionally, soilless plant growth media has been prepared from materials such as peat, pine bark, cocoa fiber, perlite, and vermiculite. However, growth media can be prepared from any materials that can provide good physical condition and supply enough nutrients for plant growth. Coal combustion products (CCPs) also have a great potential to be used in production of greenhouse and nursery media.

Compost materials and organic wastes, if mixed with CCPs, can provide good physical condition for seed germination and plant growth as well as supply plant nutrients. Most CCPs have alkaline properties while most organic composts are acidic in nature. Growth media prepared by mixing the right proportion of CCPs with organic composts will help provide proper pH characteristics as well as supply essential plant nutrients, thus supporting good plant growth. Several studies have proposed the use of several waste materials, listed above, as components for creating soilless media to grow plants in greenhouse and nursery conditions such as horticultural wastes (Stabnikova et al. 2005), biochips, sludge and fly ash (Tan et al. 2004), pruning waste compost (Benito et al. 2005), bottom ash and compost (Bardhan et al. 2008; Black and Zimmerman 2002) and FGD-residues (Sloan et al. 1999). Studies have demonstrated the successful utilization of papermill sludge in potting media, after composting for production of container-grown greenhouse and nursery crops (Chong and Cline 1993; Chong et al. 1987; Cline and Chong 1991). Campbell et al. (1995) suggested that the compost should be leached to reduce salt levels and have its pH lowered before it could become suitable for use in soilless potting mixes.

In Canada, Ensyn Technologies has developed a Rapid Thermal Processing (RTP) reactor which heats biomass to an extremely high temperature (400–900 °C) for 0.5 s at atmospheric pressure with no oxygen (Rodden 1993). RTP is also called fast cracking and is similar to the catalytic cracking process used by the oil industry. The rapid heating of the biomass cracks the chemical bonds and produces a liquid bio-oil. Rapid cooling prevents the completion of chemical reactions. The feed stock can vary: pulp sludge, wood waste, rice husks, agricultural residue. The bio-oil created from the process has been used as a fuel oil substitute. Destructive distillation as a resource recovery process for solid waste was evaluated during 1982–1984 at Marcel Paper Mills, Elmwood Park, New Jersey (United States) (FioRito 1995). The results indicate that the process is environmentally friendly and has the ability to provide substantial energy savings utilizing organic solid waste as its sole source of fuel. The technology is able to fractionate the biomass content of municipal and industrial wastewater sludge to a combustible gas and inert char in an environmentally safe manner. Full scale operation of the process was carried out on sewage and deinked paper mill sludge at installations in California and New Jersey (Weigand and Unwin 1994).

Conversion to fuel components: Levulinic acid has been found to be economically produced from paper sludge and converted into an alternative fuel component called methyltetrahydrofuran which can be used with ethanol and natural gas liquids to create a cleaner-burning fuel.

The expense of solids disposal could be eliminated by destroying the microorganisms in the excess secondary sludge and recycling the material through the treatment process. Springer et al. (1996) used a simple mechanical device- a kady mill to breakdown the microorganisms in the excess sludge allowing all of the material to be recycled to the treatment process. The kady mill combines the effects of high shear and temperatures, both of which are required for efficient cell destruction. Based on 60 days of operating data, it was found feasible to operate an activated sludge plant in extended aeration mode by recycling sludge that has been lysed in a kady mill. This process could be an alternative wastewater-treatment system for use in the pulp and paper industry. The system would be most suitable for use in mills operating well within EPA permit discharge limits for BOD. This system operated with an average COD-removal efficiency of 80 %, compared with 87 % removal for the conventional system. Both systems operated with an influent COD of 260 mg/L. The sludge-lysis-and-recycle process operated free of bulking problems. This process appears to be an economically attractive alternative to conventional treatment if higher BOD values can be accommodated.

Hammond and Empie (2007) have reported that Secondary waste water sludge can be added to the black liquor gasification process at a paper mill to produce a combustible fuel gas. The gas is fed to a combined cycle boiler plant and turbogenerator system to generate electricity.

A method of treating paper mill sludge treatment as raw material for the manufacture of animal bedding won a National Recycling Award for EnviroSystems, Cheshire, UK (Anon 2005). The sludge is dried down to 90 % dry material and broken in small pieces, and then is heat treated. The finished product is called EnviroBed and is being used as bedding for 50,000 dairy cows in the UK. Sludge from Bridgewater Paper and Shotton Paper is being processed at EnviroSystems plant in Cheshire. A second plant at Brent Pelham, Hertfordshire, is being supplied by material from Aylesford. EnviroSystems is looking for additional supplies of suitable paper crumble, with 40–45 % organic matter or above and without a high moisture content.

The wastewater sludge of Neenah Paper, Neenah, WI, United States, is recycled into useful forms, including electrical power and glass aggregate (Anon 2004). 5,000 tpy of paper sludge are recycled using a system installed by Minergy Corp, also located in Neenah. Solids are melted in a glass furnace in which organic compounds are destroyed. The inorganic mineral waste exits the furnace as liquid glass which is used in the manufacture of floor tiles, abrasives, roofing shingles, asphalt and decorative landscaping materials. Via a steam generator furnace heat produces electricity which dries the wastewater solids. The recycling process provides many environmental benefits, in Neenah Paper's case preserving green space and reducing landfill use. The company has developed an online tool for individuals and businesses to calculate the environmental benefits of using recycled paper.

Oxycair is an innovative treatment technology developed to treat various types of waste waters, which has been shown to generate substantial savings over conventional treatment costs (Gagnon and Haney 2005). The technology uses patented processes, is based on concurrent physical mechanisms taking place within multiple reactor vessels and uses no chemicals. The destructive mechanisms include:

- Physical destruction
- Thermic stabilisation
- Air supersaturation

Table 6.16 Composition of lime sludge

Element	%
Calcium carbonate	>95
Sodium oxide	<0.2
Silica oxide	<0.2
Aluminium	<0.5
Iron oxide	<0.5
Sodium sulphate	<0.01
Calcium oxide	<0.5

Based on Milovidova et al. (2010)

- Oxidation
- Explosive decompression
- Cavitation
- Microbubble oxidation

The technology has been tested at both laboratory and industrial scale, transforming excess sludge stream into a nearly sterile stream rich in dissolved oxygen and the nutrients and micronutrients contained in bacterial cells. This stream can be returned directly to the bioreactor as a nutrient supplement. Oxycair is a service provided by WR3 Technologies Inc, Canada.

Almost all Kraft-pulping mills are equipped with a system for lime recovering from lime sludge. Amount of lime sludge generated is 400 kg per tonne of pulp produced. Recovering is done in a limekiln that is a rotating drum kiln. Main reaction of the process is endothermic calcium carbonate decomposition. Heat consumption for lime sludge calcination is 10 MJ per 1 kg of calcined lime (Milovidova et al. 2010). Temperature of the process is about 1,100–1,300 °C, however optimal temperature of the process depends on inert additives. Inert additives, especially silicates are melted at high temperatures and, thus, generate glass-like coating causing problems when lime is slaked (Nepenin 1990). Moisture content of lime sludge fed should not exceed 55 % and content of Sodium oxide should be less than 1 %. Otherwise, rings of lime will be generated on the kiln inner surface causing technological problems. Composition of lime sludge fed into the limekiln is given in Table 6.16 (Nepenin 1990). During the caustisizing and recovering, some part of calcium oxide is lost. To cover the losses of calcium oxide in the system, limestone is used. Amount of limestone used ranges from 10 % to 15 % of lime sludge fed into the process (Milovidova et al. 2010). Limestone is used since about 52–53 % of limestone is calcium oxide that is required for the process. As of 2013, there was no scientific article available about deinking sludge utilization in drum kiln. Nevertheless, deinking sludge is expected to be rich in calcium carbonate coming from paper filler and, thus, to be appropriate for the process. Content of calcium oxide that is generated as a result of calcium carbonate thermal decomposition is shown in Table 3.7 (Chap. 3).

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Chapter 7
Examples of Pulp and Paper Mill Waste Implementation

Abstract Examples of Pulp and Paper Mill Waste Implementation are presented. Several Pulp and Paper mills – April, Inc. mills, Cartiere Burgo Verzuolo, Italy, Cartiere Burgo Mantova, Italy Jamsankosken Voima Oy, Finland; Aanevoima Oy, Finland; Vamy Oy/Vattenfall Oy, Finland; Modo Paper AB Husum; Sweden; Monsteras, Sweden; Elektrocieplowni Ostroleka, Poland; Metsa-Serla Oy Simpele, Finland; Oy Metsa-Botnia Kaskinen, Finland Boise mills, The Catalyst Paper mill, International Paper, Neenah paper, Nippon Paper Group, Norske Skog's modern mills, Stora Enso, Parenco paper mill, Duluth Mill, Minnesota Paper mill. Metsäliitto Group mills, Nippon Paper Company, VAR (a recycling company) – are implementing a number of waste stream reduction and re-use initiatives.

Keywords Waste stream reduction • Pulp and paper mill • Waste management • Soil improvement • Composting • Energy recovery • Steam • Electricity • Glass aggregate • Biofuel • Incineration • Deinking sludge • Waste water treatment sludge

State of the art waste reduction techniques employed in the Pulp and Paper Industry worldwide are presented below (Bird and Talbert 2008):

- April, Inc. mills is burning sludge wastes in the power boiler (April 2006).
- Some European pulp mills which burn some portion of pulp sludge are –Cartiere Burgo Verzuolo, Italy; Cartiere Burgo Mantova, Italy Jamsankosken Voima Oy, Finland; Aanevoima Oy, Finland; Vamy Oy/Vattenfall Oy, Finland; Modo Paper AB Husum; Sweden; Monsteras, Sweden; Elektrocieplowni Ostroleka, Poland; Metsa-Serla Oy Simpele, Finland; Oy Metsa-Botnia Kaskinen, Finland (CANMET 2005).
- Boise mills is using byproducts to make compost mixture, cement additives, potting soil, landscape bark, and roofing shingles (Boise 2006).
- At Boise mills sludge that settles in the waste treatment process is dewatered and burned as an alternate renewable energy source to create steam (Boise 2006).
- The Catalyst Paper Recycling Division sent 31,000 tons of residuals – inert carbon ink and paper fiber – to customers instead of landfills in 2006. Customers use the residuals as a growing medium for turf (Catalyst 2006).

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- At International Paper, goal for bleached mills is to achieve a fiber loss rate of less than 1 % and for unbleached mills to achieve a loss rate of less than 12 % (International Paper 2006).
- In Metsäliitto Group mills, fiber sludge from the paper and board mills and ashes left after energy production are used for soil improvement as such or composted at Metsäliitto Group mills (Metsäliitto 2006).
- Some 5,000 tons of sludge from all of Neenah's paper brands is converted to steam, electricity, and glass aggregate every year. The primary objectives of this recycling process is to reduce the load on landfills, which carries out a corporate environmental directive. Neenah then purchases the steam back to dry paper during manufacturing and also to heat its mill. The company projects that using this "green steam" will reduce its natural gas consumption by 80 % annually (Neenah 2006).
- At Nippon Praper Group, paper sludge ash was used effectively in roadbed construction and soil improvement (2006).
- Nippon Paper Group has observed that due to the trace amounts of heavy metals contained therein, untreated paper sludge ash cannot meet soil environmental standards. Nippon Paper Industries' Kushiro mill has been developing hydrothermal solidification equipment that crystallizes and seals in heavy metals that are contained in paper sludge ash. Verification testing of the equipment began in fiscal 2006 to prepare for actual operation. Products that have gone through the granulation and hydrothermal reaction are lightweight, porous, and have good drainage properties. Taking advantage of these properties, such products are to be used as soil improvement agents (Nippon Paper Group 2006).
- Norske Skog's modern mills utilize by-products, such as sludge from waste water treatment and deinking plants, and other organic waste from the production process as biofuel for thermal energy production (Norske Skog 2006). Econova has been working at Stora Enso Skoghall, Sweden since the year of 2002. Econova utilizes the waste products from Skoghall through sun and wind drying (since 2003) and composting (since the beginning of the '1990s). The two recycling areas are Värhult, an old landfill (14 ha) and Vidö Gård, a mill site (7,5 ha). Sixteen thousand ton 30 % DS of primary sludge goes to the recycling area Värhult for Sun and wind drying through the Econova method, giving 65 % DS (4,800 ton DS) back to the bark boiler replacing conventional dry woodchips. Thirty-five thousand six hundred ton ~25 % DS of chemical sludge goes to the recycling areas Värhult and Vidön (within the factory area) for composting, giving an important soil component for Econova's soil production. Three thousand six hundred ton bioash are used for construction. Two thousand nine hundred ton of bark sludge goes to the recycling areas Värhult and Vidön for composting, giving an important soil component for Econova's soil production.
- Sludge and ash in Australia and Asia are sometimes used for soil improvement in agriculture at Norske Skog mills (2006).
- Stora Enso has worked on improvements in waste water treatment plant nutrient control. Nitrogen and phosphorus are added as nutrient sources for the biological organisms in the waste water treatment process (Stora Enso 2006).

- Coarse reject from the screening phase of paper recycling process is well suited for energy recovery due to its high calorific value (Dehue et al. 2006). In Parenco paper mill at Netherlands a bio-boiler is used to incinerate all paper waste streams (deinking sludge, primary sludge, secondary sludge and rejects). After removing ferro metals from the paper recycling rejects they are crushed and mixed with other components (such as wood and sludge) and stored. The sludges (deinking, waste water treatment sludge) are first mixed and pressed to obtain 50–60 % dry matter and then fed to the boiler. The boiler can handle up to 390 tonnes per day of dried solid fuel. The installation can handle 30 ton/h (approximately 240,000 ton annually). The boiler produces max. 48 MW thermal and max. 15 MWe electricity by using a back pressure steam turbine. The low pressure steam (three bars) is consumed internally in the process. After incineration ashes are leftover (about 30 % of original volume). These can be landfilled or used for production of cement, asphalt or ground improvement. The investment in the incineration installation by Parenco was, when it was built (2003–2004), around 35–40 million euro. The payback time varies from 3 to 10 years depending on

  1. The type of waste streams used
  2. Any forms of subsidies
  3. Savings on energy/disposal costs

  In the case of Parenco the annual savings from reject incineration are around 800,000 euro (16,000 rejects per year). The ashes are given to the cement industry. Currently this still requires a disposal fee, but Parenco has indicated that this may change from costs to profit in the future

- Stora Enso combusts waste water treatment and de-inking sludge in Hylte Mill's newly rebuilt biofuel boiler. The Duluth Mill has an increased use of de-inking sludge for daily cover at municipal landfills (Stora Enso 2006).
- Boiler ash has been applied in road construction and concrete brick manufacture from APRIL, Inc. mills (2006).
- Wood ash waste from the boilers at the Boise International Falls, Minnesota, paper mill is spread on local farmland to improve soil pH (Boise 2006).
- Wood ash is used as a fertilizer at Metsäliitto Group mills (2006).
- Nippon Paper Company continues to develop various technologies, including hydrothermal solidification technology, so as to find new uses for incinerated ash at each mill (Nippon Paper Company 2006).
- Stora Enso uses all boiler ash from Anjalankoski Mill in road construction projects (Stora Enso 2006).
- The company CalciTech has developed a process for the production of synthetic calcium carbonate (SCC), an advanced form of precipitated calcium carbonate (PCC) (www.calcitech.com). The new process is able to separate paper sludge ash into an ultra pure calcium carbonate and a form of metakaolin. According to CalciTech the SCC recycled mineral has a positive influence on the gloss, brightness, opacity and printability of the coated paper end product. This is due to its narrow particle size distribution compared to PCC or GCC and

its high brightness. A small scale plant located in Eastern Germany currently produces samples for customers interested in testing the SCC in their products (www.calcitech.com).

- The VAR a recycling company in the Netherlands has together with a partner developed a technology that can separate the fibre and plastic fraction of coarse rejects from the paper industry (CEPI 2011). The separated fibres can be reused in board production process thereby saving transportation costs and feedstock costs. The plastic fraction can be incinerated with energy recovery, although increased plastic recycling systems in many countries have increased the development of higher added value applications for recycled plastics. Success of the technology depends on the application possibilities of the recovered fibre and the willingness of the paper companies to use these fibres. The technology has had extensive testing on several machines at VAR using the rejects of different paper mills. The machine has also been tested for other streams that could potentially provide feedstock for the paper industry. According to the calculations made by VAR roughly 95 % of the fibres can be isolated from the reject stream.
- At APRIL, Inc. mills, the boiler uses black liquor recovered from the manufacturing process and bark and rejects (APRIL, Inc. mills 2006).
- At Boise mills, sources of self-generated energy, such as wood wastes, pulping liquors, and hydroelectric power, provided 63 % of total energy requirements in 2005 (Boise mills 2006).
- In pulp production at the Metsäliitto Group mills, the chemicals in the cooking liquor are recovered for reuse, and the lignin dissolved in the cooking liquor is used for energy production (Metsäliitto Group 2006).
- At APRIL, Inc. mills screen rejects are used for second grade paper production (APRIL, Inc. mills 2006).
- Sources of self-generated energy, such as wood wastes, pulping liquors, and hydroelectric power, provided 63 % of total energy requirements in 2005 at Boise mills (2006).
- Wood waste from wood products plants and paper mills is burned as fuel at Boise mills (2006).
- Most of the fiber Catalyst uses consists of residuals from British Columbia sawmills – chips, shavings and sawdust. The company also uses poor quality softwood logs that are defective or otherwise unsuitable for lumber manufacture, and deinked pulp recycled from old newspapers and magazines (Catalyst 2006).
- International Paper wood products mills frequently sell shavings and bark to companies that use these raw materials as a greenhouse gas neutral substitute for natural gas and coal (International Paper 2006).
- At Metsäliitto Group mills, by-products, such as woodchips, sawdust and bark, are used as raw materials for chipboard and pulp production or in heat generation (Metsäliitto Group mills 2006).
- Most of the wood that is not converted into products is utilized either in energy production at Metsäliitto's own production units or as biofuel sold outside the Group (Metsäliitto Group 2006).

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Chapter 8
Future Prospects

Abstract It is necessary to continue research on different applications of wastes, while taking into account the environmental and economical factors of these waste treatments. Pulp and Paper mills are already implementing a number of waste stream reduction and reuse initiatives. Some additional considerations for greater participation include: Resource savings and resource efficiency; Substituting fossil energy sources for waste based sources and on site composting can be a potential source of carbon credits for mills participating in emerging carbon markets; Waste products can be used to support mills' infrastructure with such items as road building components, soil amendments, fuel for electricity, and construction materials. Future research by government and non profit organizations as well as the industry will reveal the extent of potential cost savings associated with these options for waste stream. However, the minimisation of waste generation still has the highest priority.

Keywords Pulp and paper mill • Waste stream reduction • Resource saving • Resource efficiency • Composting • Carbon credit • Soil amendment • Fuel for electricity • Construction material • Minimisation of waste generation

8.1 The Future

Pulp and paper industry produces enormous quantities of solid waste which presents huge environmental burden (Monte et al. 2009). Appropriate management of this waste is most crucial task for modern pulp and paper industry. The pulp and paper industry has conducted or sponsored most of the research on identification and evaluation of management options for paper industry solid wastes. This has been common research topic for more than 50 years.

The waste is very diverse in composition and consists of rejects, different types of sludges and, in case of on-site incineration, ashes (Monte et al. 2009; Bird and Talbert 2008; Kay 2002, 2003; Deviatkin 2013; Kujala 2012; Likon and Trebše 2012; Bajpai et al. 1999). The production of pulp and paper from virgin pulp generates less waste and the waste has the same properties as deinking waste, although with less inorganic content. Within the European Union several already issued and other, foreseen directives have great influence on the waste management

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strategy of paper producing companies. Through legislation, the landfill option is restricted, although it has not phased out on-site landfills. Due to the large quantities of waste generated, the high moisture content of the waste and the changing composition, some recovery methods, are simply too expensive and their environmental impact uncertain. The thermal processes, gasification and pyrolysis, seem to be interesting emerging options, although it is still necessary to improve the technologies for sludge application. Other applications, such as the hydrolysis to obtain ethanol has several advantages but it is not well developed for pulp and paper sludges. Therefore, at this moment, the minimisation of waste generation still has the highest priority.

Based on literature review and chemical analysis of deinking sludge, it is proven that sludge most successfully can be utilized in cement, ceramics, lightweight aggregate and fiberboard production. Sludge can be potentially utilized in cement mills and brick mills (Deviatkin 2013; Kujala 2012). Even though individual studies have proved that deinking or paper mill sludge is a viable reinforcement, additive or composite in various applications, all sludges vary in composition and therefore their suitability to raw material must always be tested with real life trials. To enable trials, co-operation between industry fields must be established. Companies should take initiative and actively find nearby operating producers that use raw materials present in the sludge. Successful large scale trials between industrial operatives would be a significant step towards improved material efficiency. Based on the quantity of research data pertaining to sludge gasification, it is evident that there is significant interest around the globe in developing this technology to commercial scale (US EPA 2012). Although there are many options when it comes to novel methods of sludge disposal and utilization, gasification is currently receiving the most attention. Other novel technologies, such as, super critical water oxidation, which are less mature may be suitable options, but not enough research on these technologies has been performed. With a handful of gasification systems in the final stages of development, only a leap from pilot scale to commercial scale is needed, with more technologies following closely behind.

Literature detailing the achievements of some paper sludge processing techniques suggests a promising outlook. A rational development program that embraces waste reduction at source (example, reduced fiber losses to drain) in conjunction with a comprehensive examination of paper sludge uses is needed. While national programs are useful, emphasis needs to be placed on local needs that exemplify the proximity principle and ‘virtuous cycling of resource’. Serving local needs will reduce transport costs sufficiently to make the process viable.

Secondary sludge waste management issues are a big challenge especially with the implementation of more stringent environmental legislation. Typical post-treatment methods for secondary sludges include incineration, pyrolysis, gasification, direct liquefaction, super critical water oxidation and anaerobic digestion. The operating conditions (temperature, pressure, atmosphere and products, etc.) vary among the methods. For example, incineration, gasification and SCWO methods utilize air or oxygen while the remaining methods are conducted under oxygen depleted or anaerobic conditions. Incineration, pyrolysis and gasification operate at high temperatures, while these methods differ in the objective products. Incineration aims to produce heat and steam/electricity, pyrolysis targets a high yield of oil, and gasification favors production of gas. The greatest sludge volume reduction (over 90 %) can be achieved with the high-temperature methods including incineration, pyrolysis and gasification, which is advantageous as it effectively reduces the physical amount of sludge for disposal. The major disadvantage for these high-temperature processes is their lower net energy efficiency for the treatment of secondary sludge containing very high content of water (98–99 %), resulting from the need of the energy intensive operations of dewatering/thickening and complete evaporation of the water in the sludge. In contrast, the other three treatment methods, i.e., direct liquefaction, SCWO and anaerobic digestion, operate at a relatively lower temperature and more importantly without the need of dewatering /thickening and complete evaporation of the water in the sludge. Accordingly, these methods are more promising for the treatment of secondary sludge from the standpoint of energy recovery. Proteins have been recovered from secondary paper sludges and used as wood adhesive.

Research has shown that biorefining of the waste sludge can decrease the environmental impacts of the current disposal practices significantly. This new waste reuse approach helps the industries by assisting the facilities in meeting the environmental regulatory requirements at a lower cost. Utilizing the waste sludge as filler in biocomposites, not only helps reducing the greenhouse gas emissions, but also addresses the problem of persistent plastics in the environment (Edalat Manesh 2012). This approach also helps preserving the precious resources including petroleum, minerals and forests by replacing them in the biocomposite.

Pulp and Paper mills are already implementing a number of waste stream reduction and reuse initiatives. Some additional considerations for greater participation include (Bird and Talbert 2008):

- Resource savings and resource efficiency – reducing and reusing waste can save mills money on purchased energy and material costs and reduced landfill fees.
- Substituting fossil energy sources for waste based sources and onsite composting can be a potential source of carbon credits for mills participating in emerging carbon markets.
- Waste products can be used to support mills' infrastructure with such items as road building components, soil amendments, fuel for electricity, and construction materials.

Future research by government and non profit organizations as well as the industry will reveal the extent of potential cost savings associated with these options for waste stream reuse (Bird and Talbert 2008):

Full-scale experience suggests that the viability of alternative management strategies primarily depend upon four factors (Weigand and Unwin 1994):

- Technical feasibility
- Cost
- Available markets
- Potential liability

The relative significance of these factors varies depending on mill type, mill location, waste type, and company business strategy. There has been considerable interest in some of the solid waste management alternatives as stated above. The interest in these particular waste management opportunities probably relates mostly to their potential for using significant amounts of sludge.

There are good prospects for recovering usable fibre and fillers from paper sludges in paper making, and, therefore, this area warrants further assessment for sludges from a variety of different mills and product lines. Wet air oxidation processes have been extensively examined in this regard, but have not been adopted widely due to costs. In light of current economic and environmental circumstances, it is prudent to revisit potential uses for this technology as well as cleaning and screening technologies which have resulted in commercial scale recoveries of fibre and filler from some sludges. The use of paper sludges in cementitious product applications warrants considerable attention. Although initial bench-scale testing is required for many of the prospective applications, larger-scale pilot studies are required to establish the practical and technical feasibility of each sludge use option.

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