An Advanced Course in Nuclear Engineering

Shinya Nagasaki
Shinichi Nakayama *Editors*

Radioactive Waste Engineering and Management

Springer

An Advanced Course in Nuclear Engineering

Since the beginning of the twenty-first century, expectations have been rising for nuclear power generation in the world because the need for a safe and stable energy supply is increasing against the background of global environmental issues and the depletion of oil-based energy sources. The situation is calling for the development of human resources with advanced knowledge and techniques of nuclear energy. The role of nuclear energy remains unchanged in the world even after Fukushima Daiichi Nuclear Power Plant accident. Needs for education and human resource development of nuclear professionals who understand and manage the nuclear power have increased worldwide.

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We would like to extend our gratitude to all those who have kindly taken the time to contribute to or cooperate in the creation and publication of this book series.

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Foreword

In the aftermath of the Fukushima Daiichi nuclear accident, the need for sciences of nuclear waste and material management is diversified as well as increasing. Without a proper long-term solution for managing a huge amount of soils and other off-site environmental matters contaminated by the radioactive substances released from the accident as well as managing a variety of on-site radioactive wastes arising from dismantlement of damaged reactors, the accident clean-up would never come to an end and the nuclear renaissance for globally and sustainably using nuclear power to produce energy would never come true, either.

Publication of this book, therefore, is truly timely. In particular, it could be tremendously useful as a textbook, since it is defined as a series of "An Advanced Course in Nuclear Engineering," edited for the international graduate student course at the University of Tokyo.

There are a couple of unique features observed in the book. Firstly, all the authors and co-authors are those who actively work at the forefront of radioactive waste management. Primarily because of that, what is delineated here is well balanced between theory and practice, although many textbooks tend to pay much attention to theoretical aspects.

Secondly, it embraces a wide spectrum of scientific areas related to the issue of radioactive waste management, which is not limited to technical subjects but is closely linked with the relations with society. It provides a scientific basis for facilitating a profound understanding of the socio-technical aspects of the issue.

By and large, radioactive waste management encompassing technology and society used to be treated with lower priority, compared with nuclear physics, nuclear reactor engineering, nuclear material sciences, and so on. The time has come, however, to face the challenge with the greatest emphasis, because now people do recognize that a key to success of peaceful use of nuclear energy or more broadly of the application of society-linked technology is whether human beings are capable of managing radioactive waste. My hope is that this book will stimulate younger generations to explore the possibility of resolving such challenges, not merely in nuclear areas but more generally.

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Foreword

The history of issues surrounding radioactive waste is relatively short and the area of study continues to expand. It is also an inexpiable area filled with contradictions in that no countries in the world so far have attained geological disposal of high-level radioactive waste, which is the most important and difficult task, while enjoying the benefits brought by nuclear power and radiation use.

Issues of radioactive waste cannot be discussed without referring to the extensive list of related laws and regulations and carries within itself the issues of social, economic, political, environmental issues as well as intergenerational ethics. The issue also requires discussions with a view of the future in hundreds or hundreds of thousands of years, and because of this time scale it contains unavoidable uncertainties.

I myself have faced difficulties in real life since the time I was teaching at a faculty of engineering in terms of thoroughly covering and discussing the issues of radioactive waste, even with a limited focus on engineering, that are characterized by the aforementioned complexity.

Radioactive Waste Engineering and Management has now been published and describes this multifaceted issue in a detailed and informative manner beyond the entitled focus on regular engineering interest by contributions made by up-and-coming experts by way of fine and careful assignment and coordination.

The book is ambitious in that it endeavors to cover the issues from decommissioning to clearance, reaching to geological disposal of high-level radioactive waste. It is pleasant to feel the young and energetic momentum and determination of the contributors.

As mentioned, simply by recognizing the fact that the most significant issue of radioactive waste disposal, namely, geological disposal, has been left unresolved, and the fact that all aspects of radioactive waste issues are closely linked to societal and environmental issues among others, it is possible that the contents of and approaches to the items covered in this book will be subject to change year by year. Therefore, I would think the contributors to this book will be required to continue revision, expansion, and enrichment of the textbook at an appropriate timing. This should require continuation of substantial efforts, and it is my wish that such endeavors shall be accomplished.

This book presents the issues of radioactive waste from multifaceted and comprehensive points of view, and there is no doubt that it will be an extraordinarily excellent textbook for readers from a wide range of backgrounds. It is my sincere hope that the younger generations will, through this textbook, delve into the challenging domain of radioactive waste issues or make use of the book for resolving the issues of the individual reader.

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Kunio Higashi

Preface

Responsible management of radioactive wastes is an important issue for each and every country that makes use of nuclear power and radiation and radioactive isotopes for industry and medicine. Since the end of the twentieth century, the issue of intergenerational ethics in the development and use of nuclear power and materials has been recognized as essential to avoid passing a negative legacy to future generations as a result of the present generation's enjoying the benefits of a convenient life without thinking responsibly about the burden that such a life may place on those who come after.

The twenty-first century is expected to see an increase in the deployment of light water reactors, particularly in Asia, and an essential part of any forward-looking agenda will certainly be the issue of radioactive wastes. While developing countries look at nuclear power to address their growing demand for energy against dwindling resources, it is important to understand that the future development of nuclear power will depend greatly on our success in establishing the required technology for effective management of radioactive wastes. The use of nuclear energy in the twenty-first century is premised on the realization of reasonable radioactive waste management that can ensure public safety, security, and reliability.

While most people are aware that nuclear power facilities generate wastes, research and medicine are other sources where the use of radiation and radioactive isotopes has become routine. Added to this is the waste generated when facilities that employ radioactive materials are dismantled after closure. It is the responsibility of our generation to establish safe and effective systems for the handling of such radioactive wastes for today and for future generations.

This book is designed to provide everyone with an interest in radioactive waste issues, including students and individuals involved in engineering and public administration, with the scientific foundations that support radioactive waste management at a graduate level. This book is also intended to help readers better understand the role of mass transfer and chemical equilibrium theories in the clearance and radioactive waste handling processes.

In brief, the items included in *Radioactive Waste Engineering and Management* are explained in the following. While all processes in the handling of radioactive wastes, from generation to disposal, are often comprehensively described as radioactive waste management, the definition of management is often interpreted as the physical and chemical processing of generated wastes to delivery to a disposal facility and closure of that facility. This thinking is most likely the result of people's tendency to limit the range covered by their direct control. However, radioactive waste management should not be limited in this way. Instead, it should include physically and conceptually wider areas that cover safety regulation concepts and social involvement.

Dismantling of nuclear reactors and nuclear-fuel-cycle facilities has become an emerging interest in the nuclear industry. The generation of radioactive wastes by this action at the end of a facility's lifetime is not the only area of concern. The decommissioning method and the regulatory system for clearance and exemption of very low-level radioactive wastes have a profound impact on the amount and processing of secondary wastes. The management of the wastes exhibits complex coupling among technologies, regulations, and society's acceptance of those clearance and decommissioning systems. This book addresses bases to understand the dismantling and decommissioning of nuclear facilities.

In addition, it is essential to establish the safety of radioactive waste disposal, including deep geological disposal, over a period of time that extends beyond a few generations. How can we prove the safety of radioactive waste disposal with the science and technology that we now possess? What criteria should be provided for the regulations? Also in this issue, we observe complex coupling among technology, regulations, and society in an unprecedented long-time scale. Seeking engineering needed to realize it may bring us to a new engineering horizon completely different from what we now know. This book includes comprehensive coverage of relevant laws and regulations, as well as all technological processes ranging from facility decommissioning and conditioning wastes to long-term safety assessment for final geological disposal. As one of the waste-conditioning technologies, for which active research and development have been performed, roles and effects of partitioning and transmutation are also discussed.

The concept of radioactive waste disposal has developed through extensive and wide-ranging international discussions. Because future use of nuclear energy must be based on safe and rational radioactive waste management, it is necessary to prioritize the establishment of what it should be, and those involved in this field will be required to possess a thorough understanding of the overall picture. We as the authors of this book, therefore, did our best to systematically summarize such knowledge from the perspective of engineering. It is our hope that the readers of this book will play a responsible role in radioactive waste management.

While the issues of radioactive waste management associated with nuclear weapons production are important for nuclear-weapon states, such issues are not completely unrelated to non-nuclear-weapon states under a nuclear deterrent, including Japan. We have decided not to cover those weapon-related issues in this book.

The Fukushima Daiichi accident in March 2011 crippled four nuclear reactors and generated an enormous amount of unprecedented radioactive wastes both on-site and off-site of the Fukushima Daiichi plant. Many issues and difficulties are observed to have similar coupling that we have discussed in this book, i.e., technology, regulation, and society. It is clear that we need to create innovations to resolve this long-lasting challenge of decommissioning and radioactive waste management by developing not only radioactive waste engineering but also a public decision-making process that can actually result in socially agreeable solutions and management. This edition, which is a translation of the original Japanese version published before the accident, does not explicitly address Fukushima issues and includes minimal update where appropriate, but the authors believe that the contents of this book provide the foundation essential for such future development. The authors hope that they will have an opportunity to include achievements since the Fukushima Daiichi accident in a future edition of this book.

Hamilton, ON, Canada
Tokai, Japan
July 2014

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Chapter 1
Radioactive Waste Management

Shinichi Nakayama

1.1 Principles of Radioactive Waste Management

All kinds of activities associated with radioactive wastes generated as a result of nuclear utilization are often collectively referred to as "radioactive waste management." This term is used in a broader sense than a mere series of technical processes to convert generated wastes into waste forms for storage and disposal.

Wastes are generated through both the operation of facilities and the decommissioning of end-of-life facilities. Although only a limited amount of the wastes generated through facility decommissioning are radioactive (see Chap. 3, "Decommissioning of Nuclear Facilities"), the act still generates radioactive waste and therefore decommissioning constitutes an important part of radioactive waste management.

There are two types of radioactive waste disposal. The first type builds disposal sites within areas where people are living and monitors wastes buried there until safety concerns disappear. The second type builds disposal sites isolated from living areas because the time period required until radioactivity is sufficiently reduced is too long to keep monitoring the wastes; an example of this is geological disposal of high-level radioactive wastes. For the second type, methodologies need to be developed to assess safety of disposal for such a long time period after their closure. Partitioning and transmutation technology, for which R&D is carried out as a future technological option, is described in Chap. 2, "Generation and Characteristics of Radioactive Wastes."

The nuclear regulatory system in Japan has been changed significantly after the Fukushima Daiichi Nuclear Power Station accident in March 2011. Descriptions in this chapter have been translated from the book originally published in Japanese before the accident, with minimal update where appropriate.

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Safe management of radioactive waste involves an intergenerational equity issue. The radioactive waste management includes issues with different time scales; at least a few years from waste generation to treatment and disposal, a few decades for management after the closure of disposal sites, and even a few generations or a century when monitoring and other management activities are taken into consideration. How to manage radioactive waste safely is also an issue of intragenerational equity, because people are equally benefitting or will benefit from nuclear power generation and radiation uses, and disposal facilities will be built in their environment.

How such inter- and intra-generational equity and fairness for radioactive waste management should and could be tackled has been discussed from various viewpoints by many institutions, including international organizations, as a fundamental issue of radioactive wastes. The ideas recognized as being of prime importance were published in 1995 by the International Atomic Energy Agency (IAEA) as the nine principles of radioactive waste management (Table 1.1). These principles serve as the basis for dealing with protection levels against radioactive wastes and their effects on future generations.

Table 1.1 IAEA's principles of radioactive waste management [1]

Principle 1: Protection of human health	Radioactive waste shall be managed in such a way as to secure an acceptable level of protection for human health
Principle 2: Protection of the environment	Radioactive waste shall be managed in such a way as to provide an acceptable level of protection of the environment
Principle 3: Protection beyond national borders	Radioactive waste shall be managed in such a way as to assure that possible effects on human health and the environment beyond national borders will be taken into account
Principle 4: Protection of future generations	Radioactive waste shall be managed in such a way that predicted impacts on the health of future generations will not be greater than relevant levels of impact that are acceptable today
Principle 5: Burdens on future generations	Radioactive waste shall be managed in such a way that will not impose undue burdens on future generations
Principle 6: National legal framework	Radioactive waste shall be managed within an appropriate national legal framework including clear allocation of responsibilities and provision for independent regulatory functions
Principle 7: Control of radioactive waste generation	Generation of radioactive waste shall be kept to the minimum practicable
Principle 8: Radioactive waste generation and management interdependencies	Interdependencies among all steps in radioactive waste generation and management shall be appropriately taken into account
Principle 9: Safety of facilities	The safety of facilities for radioactive waste management shall be appropriately assured during their lifetime

The primary pillar of these principles of radioactive waste management is the ethical consideration. The IAEA stipulates in Principle 5 that "Radioactive waste shall be managed in such a way that will not impose undue burdens on future generations." Principle 5 is the fundamental guide that the current generation, which benefits from nuclear technologies, should follow in protecting future generations and the environment. It is also a major goal of radioactive waste management.

Under Principle 5 the current generation is responsible for developing technologies, building and operating related facilities, securing funds and establishing programs for radioactive waste management. Although science and technology are expected to progress in the future, it cannot be easily expected that management, requiring direct human control will continue far into the future, assuming a permanently stable society. From this perspective, Principle 5 requires that the utmost effort must be made in the belief that safety can be ensured in the future.

Principle 5 also indicates that limited actions may be passed to succeeding generations, if needed. The timing and implementation of disposal of individual radioactive waste types may depend on scientific, technical, social and economic factors such as the availability, acceptability and developmental status of suitable sites, and the decrease of radioactivity levels and heat generation during storage, as well as on social decisions based on these factors. The same applies to decommissioning. How to manage such a project over two to three decades after several decades of facility operation will be decided in consideration of the social environment for the site and future social needs regarding the use of the site.

Discussion on intergenerational equity has made progress over time. In 1995, the Nuclear Energy Agency of the Organisation for Economic Co-operation and Development (OECD/NEA) expressed the opinion [2] that current generations should take responsibility for not passing risk and burden problems and the obligation to resolve them to future generations. There is also an argument that consideration should be given to not only avoiding passing problems to future generations, but also not ignoring the possible opinions of future generations and not taking away the freedom of decision-making of future generations. If geological disposal, the disposal method chosen by current generations, is eliminating other future options, it is restricting the freedom of decision-making of future generations. This is also true for technology development. Current generations are responsible for not neglecting necessary R&D so that technological options for future generations will not be limited. In other words, the principle of intergenerational equity, which originally meant the obligation to minimize burdens on future generations, has been transformed into a concept that additionally includes the obligation to ensure future generations have opportunities equal to those of current generations with respect to decision-making on resource use and safety-related judgments.

To meet the requirement of allowing decision-making by the society of the time, geological disposal takes the approach of carrying out disposal projects step by step rather than hastily. Safety is enhanced and burdens on future generations reduced in each phase of a project, while key decision-making relies upon the generation of the time when a decision is needed. This is called the staged approach. Specific measures such reversibility and safety cases to support safety involved there are explained further in Chap. 6, “Radioactive Waste Disposal.”

1.2 Types of Radioactive Wastes

1.2.1 Definition of Radioactive Wastes

Radioactive wastes are defined as spent radioactive materials and materials that are contaminated with radioactive materials and bound to be disposed of without any further use. Radioactive wastes are generated from nuclear energy uses, typically nuclear power generation, as well as from the use of radioisotopes for medical, agricultural and industrial purposes. Under the Japanese legal framework, radioactive wastes are divided into two types: nuclear fuel materials, controlled by the Act on the Regulation of Nuclear Source Material, Nuclear Fuel Material and Reactors; and radioisotopes, regulated under the Law Concerning Prevention from Radiation Hazards due to Radioisotopes, etc. It should be noted that the final action of these materials is referred to as “disposal,” while the act of reserving materials for future use is “storage.” Strictly speaking, the term “final disposal” is redundant, although easy to understand, and “final storage” makes no sense.

Spent fuel removed from the reactor is not treated as waste in countries such as Japan, where reprocessing takes place, because uranium and plutonium are extracted from it. Residues from this extraction are high-level radioactive effluent, which will be solidified into vitrified waste. In countries such as the U.S., Sweden and Finland, which do not conduct reprocessing, spent fuel has no further use and is therefore considered as waste. Depleted uranium generated by uranium enrichment is not radioactive waste in Japan because the “Framework for Nuclear Energy Policy,” which was published in 2005 by the Japan Atomic Energy Commission (JAEC) to define programs for Japan’s nuclear research, development and use, states that this material should be stored for future use.

Radioactive wastes are generated by industries handling radioactive materials but are not called industrial waste. The Waste Management and Public Cleansing Act (Waste Management Act) defines industrial and municipal wastes as “garbage, bulky garbage, cinders, sludge, excrement, waste oil, waste acid, waste alkali, animal corpses, and other soiled or unneeded materials that are either solid or liquid (except radioactive materials and materials contaminated with them).” These wastes are clearly distinguished from radioactive wastes.

Radioactive wastes are one kind of waste containing radioactive materials; that is to say, wastes containing radioactive materials are not always radioactive wastes. Substances not regulated as radioactive materials and materials contaminated with them are not classified as radioactive wastes, in the first place. For example, radiation sources used for calibration and smoke detectors are not subject to regulation because they contain only trace amounts of radioactive materials and therefore pose only a small radiation risk. When these products are disposed of, they are not treated as radioactive wastes. When such items are not regulated as radioactive materials, they are in a state known as "exemption." An idea similar to exemption is "exclusion." Exclusion is removing materials that cannot be regulated or that are not worthwhile regulating (e.g., cosmic rays, ^{40}K in the human body) from the scope of regulation. International discussion is ongoing on whether to exclude naturally occurring radioactive materials (NORM), from the viewpoint of health science. This is because NORM includes technologically enhanced naturally occurring radioactive material (TENORM). If the discussion leads to the conclusion that NORM should not be subject to exclusion, wastes containing NORM will have to be regulated as radioactive wastes, and will eventually be disposed of as radioactive wastes unless they are freed from regulation by the clearance system (described below).

The act of freeing a regulated radioactive material or a material contaminated with radioactive material from regulation is called "clearance". The criteria for clearance are called clearance levels. Waste with radioactivity concentration below the clearance level is not classified as radioactive waste. This kind of waste is recycled into materials or disposed of as industrial waste under the Waste Management Act. In this sense, the clearance system, legislated in 2005, may be regarded as a system to permit waste from the nuclear industry to be included in the scope of non-nuclear industrial waste.

Chapter 4, "Clearance," elaborates on exemption, exclusion and clearance.

1.2.2 Classification of Radioactive Wastes

Radioactive wastes from different nuclear facilities have different properties depending on the operations taking place at each facility. Wastes are treated according to their properties and eventually transformed into a state called a waste form. Then waste forms are disposed of in a manner suitable for their respective properties. In other words, wastes generated through facility operation and dismantling change forms as they go through different stages of management. For this reason, radioactive wastes can be classified from several different perspectives. This section describes classifications at the generation and disposal stages.

1.2.2.1 Classification by Physical Properties at the Generation Stage

Radioactive wastes are in gaseous, liquid or solid forms at the generation stage and the wastes in these forms are called gaseous, liquid and solid wastes, respectively.

Gaseous wastes contain, along with gases, radioactive particulate matter, which are fine particles in the air to which radioactive materials have stuck. Gaseous wastes from reactors include exhaust gas from the steam condenser air ejector in a boiling water reactor (BWR), purge gas from the volume control tank in a pressurized water reactor (PWR), and vent gas from various components. These gases contain radionuclides such as ^{88}Kr, ^{131}I and ^{133}Xe. A typical gaseous waste from reprocessing facilities is shearing off-gas generated from the spent fuel shearing and dissolution processes, and it mainly contains ^{3}H, ^{85}Kr and ^{129}I. These gaseous wastes are subject to filtration to remove particulate matter after radiation attenuation occurs, and captured materials are solidified in a stable form for storage. For example, shearing off-gas from reprocessing is filtered with a high efficiency particulate air (HEPA) filter and captured with a silver adsorbent, and both the HEPA filter and the silver adsorbent are solidified with cement in metal drums.

Liquid wastes vary widely in terms of both solution composition and the radionuclides they contain. A typical liquid waste is effluent from spent fuel reprocessing, which consists of effluent from extraction during the first cycle of reprocessing and concentrated liquid from the acid recovery process; it includes the majority of fission products in spent fuel. This is called high-level radioactive liquid waste. After denitration, this effluent is mixed with glass material to be transformed into vitrified waste. Liquid wastes from reactors include coolant, drain, and laundry waste water. Liquid wastes are subjected to such treatment for radioactive material removal or volume reduction as coagulation-sedimentation, filtration, evaporation, and ion exchange. The residual waste is solidified; cementation is commonly used for solidification.

Gaseous and liquid wastes are treated in such a way that most of the radionuclides contained in them are transferred to solidified waste; the remaining gases and liquids are checked to ensure that their radioactivity concentration is below the regulation limit before they are released to the atmosphere or to the sea. The gases and liquids to be released are not radioactive wastes by definition.

Solid wastes vary widely as well. Diverse types of solid wastes are generated from facility operation, maintenance and decommissioning. Solid wastes made of paper, fabric, wood or polyethylene are incinerated to reduce their volume, while those made from flame-retardant materials, such as chloroethylene, rubber and plastic, and those made from incombustible materials, such as glass, earth, sand, concrete and metal, are compacted or molten to reduce their volume. Subsequently, these wastes are solidified with cement in metal drums or designated containers.

Specific practices involved in these processes are described in Chap. 5, "Radioactive Waste Treatment Technologies."

1.2.2.2 Classification by Source

Wastes generated at nuclear facilities are converted, through the described treatments, to steady-state waste forms ready for disposal. However, they still maintain the characteristics of the source facilities. These radioactive wastes in waste forms can be classified by the source as shown in Table 1.2 and identified by their characteristics.

Table 1.2 Types and volumes of solid radioactive wastes in Japan

Source	Waste name/type			Volume generated	Disposal project[a]
Reprocessing facilities	High level radioactive vitrified waste		High-level radioactive wastes	1,664 canisters in storage as of December 2009[b]; about 1,300–1,600 canisters expected to be generated annually[c]	Geologic disposal by Nuclear Waste Management Organization expected
MOX fuel fabrication facilities	TRU waste (hulls, end pieces, spent silver adsorbents, process-related concentrated liquid waste, etc.) from reprocessing facilities; combustible, flame-retardant and incombustible waste from MOX fuel fabrication facilities; returned waste from commissioned overseas reprocessing		Low-level radioactive wastes	Approx. 145,000 200-liter drums as of March 2009[b]	(Undecided)
Uranium enrichment/fuel fabrication facilities	Uranium waste (consumables, sludge, etc.)			Approx. 104,000 200-liter drums as of March 2009[c]; approx. 500,000 drums by 2050[d]	(Undecided)
Nuclear power plants	Plant waste	Relatively high-level radioactive waste (spent control rods, reactor core internals, channel boxes, etc.)		Approx. 51,000 tons by 2030[e]	(Undecided)
		Relatively low-level radioactive waste (spent ion exchange resin, filters, etc.)		Approx 220,000 200-liter drums buried as of August 2010[f]	Vault disposal in Rokkasho Village, Aomori, by Japan Nuclear Fuel Ltd. since 1992
		Very low-level radioactive waste (concrete, metals, etc.)		Approx. 13,000 tons at Japan Atomic Power Company's Tokai Power Station (output: 166 MW)[g]	Landfill disposal of waste from the Japan Power Demonstration Reactor in Tokai Village by Japan Atomic Energy Agency
Medical/research institutions	Radioactive waste generated from research, industrial and medical facilities (divided into radioisotope [RI] wastes, e.g., components, ventilation filters, medical equipment, etc. from medical and other institutions using RIs; and laboratory waste, e.g., components, ventilation filters, used test specimens, etc. from test reactors and facilities using nuclear fuel)			Approx. 560,000 200-liter drums as of March 2009[h]	To be conducted by Japan Atomic Energy Agency

(continued)

Table 1.2 (continued)

Source	Waste name/type	Volume generated	Disposal project[a]
(Reference) Industrial wastes	20 kinds of wastes from business activities that are designated by government ordinance, including cinders, sludge, waste oil, waste acid, waste alkali and waste plastics	The final landfill amount in FY 2008 was 6,349,000 tons, which accounted for 15 % of the total volume generated, or 42,629,000 tons.[i] Waste generators are responsible for disposing of their own waste	
(Reference) Municipal wastes	Wastes other than industrial wastes	The final landfill amount in FY 2006 was 21,799,000 tons, which accounted for 5 % of the total volume generated, or 418,497,000 tons.[i] Municipal governments are responsible for disposing of these wastes	

[a] For disposal methods, see Chap. 6, "Radioactive Waste Disposal";
[b] Website of the Nuclear Waste Management Organization;
[c] Website of the Ministry of Economy, Trade and Industry;
[d] Japan Nuclear Fuel Ltd. et al., "Review Paper on Uranium Waste Disposal and Clearance" (Japanese) (March 2006);
[e] Website of the Radioactive Waste Management Funding and Research Center;
[f] Website of Japan Nuclear Fuel Ltd.;
[g] Website of Japan Atomic Power Company;
[h] Materials from Japan Atomic Energy Commission;
[i] Ministry of the Environment, "FY 2007 Environmental Statistics of Japan"

During spent fuel reprocessing, nitric acid effluent, which is generated as a residue from the recovery of uranium and plutonium, is denitrified and molten with glass material for vitrification. This vitrified waste is highly radioactive and is therefore called high-level radioactive waste to distinguish it from other wastes. All other wastes are called low-level radioactive wastes. As already mentioned, in countries not carrying out reprocessing, spent fuel itself is treated as high-level radioactive waste. Wastes generated from reprocessing commissioned to the U.K. and France and returned to Japan are called repatriated wastes, and divided into two types: high-level radioactive waste and TRU waste.

Wastes generated as a result of mixed oxide (MOX) fuel fabrication and the operation and maintenance of spent fuel reprocessing facilities are characterized by contamination with transuranium elements (TRU) and called TRU waste. Among representative nuclides included in TRU wastes are transuranium and long-lived nuclides such as ^{14}C and ^{129}I. Those high-level radioactive vitrified and TRU wastes that contain long-lived nuclides are disposed of at a depth of at least 300 m. This kind of disposal is known as geological disposal, and in Japan, it is conducted by the Nuclear Waste Management Organization (NUMO).

Filters, centrifugal separators and other components used in the enrichment process during uranium fuel fabrication and sludge from the fuel fabrication process are contaminated with isotopes of uranium and are thus called uranium waste. Note that, as already mentioned, depleted uranium from the uranium enrichment process is not classified as a radioactive waste in Japan because it is considered as a resource for fuel for future fast reactors.

Wastes generated from the operation, maintenance and decommissioning of nuclear power plants are called nuclear power plant waste.

There are three types of nuclear power plant waste: low-level radioactive waste, such as spent ion exchange resin resulting from plant operation; relatively high-level radioactive waste, such as reactor core internals associated with reactor dismantling; and very low-level radioactive waste, which mainly consist of concrete and metals produced during facility dismantling.

The Low-Level Radioactive Waste Disposal Center of Japan Nuclear Fuel Ltd., located in Rokkasho Village, Aomori, is Japan's only commercially operating disposal site for nuclear power plant wastes as of 2010. Spent ion exchange resin and other low-level radioactive wastes are disposed of here.

Wastes generated from activities such as R&D using radioactive materials and radiation, improvement of the quality of industrial materials, breeding improvement and storage of agricultural products, and diagnosis and treatment of diseases are called radioisotope (RI) waste. Wastes from facilities using nuclear fuel materials, such as test reactors and hot cells in research institutes, are often referred to as laboratory waste. These wastes are collectively classified as "radioactive waste generated from research, industrial and medical facilities". Spent fuel from test reactors is included in this waste type by definition because it originates from R&D activities. Radioactive wastes generated from research, industrial and medical facilities are characterized by a variety of properties including the radionuclides they contain, radioactivity concentration, shape and materials; this is because of the diverse uses of radiation and radioactive materials for R&D. The largest source of this waste type in Japan is the Japan Atomic Energy Agency (JAEA). Since the relevant law was amended in 2010, JAEA has been engaged in disposal projects.

Nuclear industry-specific wastes that do not fall into these categories are effluent from uranium melting plants and tailings (also known as mill tailings), which are earth and sand remained after uranium extraction. Both are not classified as radioactive waste and are managed under the Mine Safety Law. JAEA's Ningyo-toge Environmental Engineering Center, which was once engaged in research in uranium extraction from ores and R&D of uranium enrichment, effluent was chemically treated to reduce contamination below the regulatory standards before being released. The tailings are stored in a heap site called the tailings dam and still monitored to prevent contamination of the surrounding environment.

Table 1.2 shows the rough volume of each type of radioactive waste generated. The amounts of radioactive waste are far smaller than those of industrial and municipal wastes. It should also be noted that due to the advancement of recycling technology, the final landfill amounts of industrial and municipal wastes have been reduced to less than 20 % of the total generation.

Japan introduced the clearance system in the nuclear field partly for the purpose of recycling and reusing materials. Recycling and reusing help to reduce the final disposal amount of radioactive wastes from nuclear facilities. Likewise, spent fuel is reprocessed in order to recycle the unused part of fuel. The reprocessing of spent fuel, through which uranium is recovered and reused, substantially reduces the disposal amount of high-level radioactive waste, compared with the case where spent fuel is simply disposed of (direct disposal). However, although effective in reducing the amount of high-level radioactive waste, reprocessing creates TRU waste. There is a constant need for developing technologies to further suppress radioactive waste generation in accordance with the IAEA's Principle 7 (Table 1.1). It is essential that the control of radioactive waste generation takes account of all types of radioactive wastes rather than looks at individual wastes.

1.2.3 Radioactivity of Radioactive Wastes

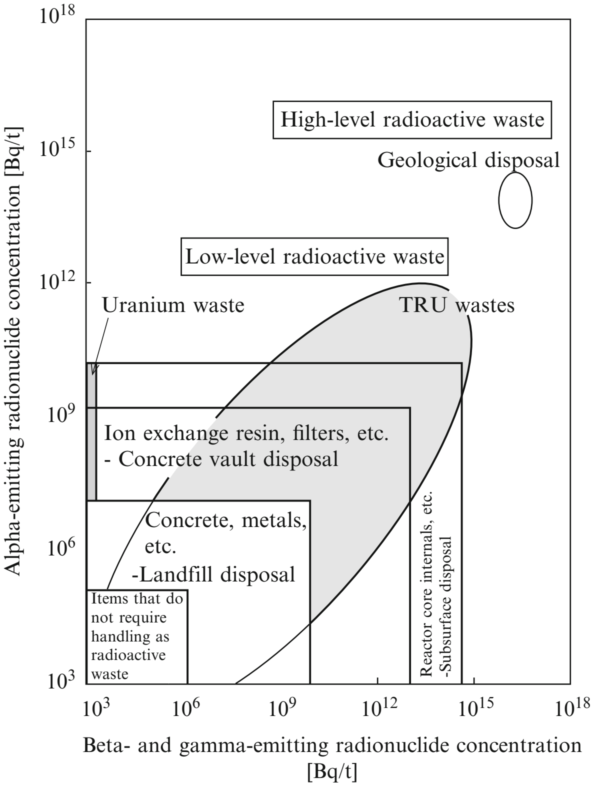
Different types of low-level radioactive wastes from various sources demonstrate different radioactivity concentrations and their distributions, depending on the characteristics of the source. Figure 1.1 shows the classification for several types of radioactive wastes by radioactivity concentration. The applicable disposal methods, such as near surface disposal without engineered barriers (hereinafter referred to as "landfill disposal"), near surface disposal with engineered barriers (hereinafter referred to as "concrete vault disposal"), subsurface disposal and geological disposal, are also indicated in the figure. The right disposal method is determined from the characteristics of the waste, especially the radioactive life of the waste and the radioactivity concentration shown in the chart (see Chap. 6, "Radioactive Waste Disposal").

The composition of high-level radioactive wastes depends on the composition of spent fuel and the specifications of treatment at the reprocessing plant (e.g., reprocessing and denitration). Although the contents of actinide elements and fission products in vitrified waste vary with the fuel burn-up, the composition stays within a specific range.

As opposed to high-level radioactive wastes, which are high-level long-lived, uranium wastes are low-level long-lived. Nuclides contained in uranium wastes at the time of generation are limited to ^{234}U (with a half-life of 250,000 years), ^{235}U (700 million years) and ^{238}U (4.5 billion years), which are naturally occurring isotopes of uranium. Unlike other radioactive wastes, uranium wastes do not contain nuclides resulting from activation or nuclear fission. For this reason, the concentration of beta- and gamma-emitting radionuclides is limited within a very low, narrow range. The concentration of alpha-emitting radionuclides is broadly distributed across a range of five or more orders of magnitude, but that of many uranium wastes is low, at a level similar to or below the concentration of naturally occurring alpha-emitting radionuclides. However, due to factors such as long lifetime, which means radioactive decay can hardly be expected, and the occurrence of radon and other progeny nuclides, uranium wastes should be addressed in a different manner than other types of wastes when clearance levels are set or safety assessment of disposal is performed.

The radioactivity concentration of nuclear power plant waste, TRU waste and radioactive wastes from research facilities (not indicated in Fig. 1.1) is distributed

Fig. 1.1 Concept of classification of radioactive wastes by radioactivity concentration



widely. Among nuclear power plant wastes, those with relatively high radioactivity (e.g., reactor core internals, spent ion exchange resin) and those with very low radioactivity (e.g., concrete, metals)—known as very low-level radioactive waste—are disposed of by different disposal methods according to their radioactivity level. By contrast, TRU wastes have long radioactive lifetime because of the various radionuclides contained in them. TRU wastes are sorted by characteristics and disposed of by rational methods suited to the waste characteristics, including geological disposal, subsurface disposal and landfill disposal.

The entry “Items that do not require handling as radioactive waste” in Fig. 1.1 indicates wastes below the clearance limit. Wastes that have been judged not to be radioactive wastes (i.e. non-radioactive waste, see Sect. 2.7, “Waste below the clearance level” and 4.1.2, “Concepts similar to clearance”) from facility usage records also fall within this category. As of 2010, items constituting cleared wastes and non-radioactive wastes are limited to the metals and concrete containing only short-lived nuclides that are generated at nuclear power plants.

1.3 Radioactive Waste Engineering

1.3.1 Radioactive Waste Treatment and Disposal Processes

Now that an overview of radioactive waste types has been provided, this section describes the process flow from generation to disposal. As Fig. 1.2 shows, the fundamental steps are pretreatment, treatment, conditioning and disposal. "Treatment" as mentioned in the phrase "treatment and disposal" refers to three processes: pretreatment, treatment and conditioning. The sources of generation are general activities involving nuclear energy and radiation uses, as just shown in the previous section. Gaseous, liquid and solid wastes are generated here.

Pretreatment is a process to select the optimal treatment method. It refers to such activities as waste collection, separation, chemical component conditioning, and decontamination. Gaseous and liquid wastes are decontaminated, and their components with sufficiently reduced radioactivity concentration are released to the atmosphere or to the sea. Non-radioactive wastes and cleared wastes are separated at this stage. This separation can be regarded as a pretreatment process. As a matter of course, some wastes, like high-level radioactive effluent, obviously have such high radioactivity levels that they cannot be subject to judgment based on usage records or to clearance verification. Among all wastes generated, those remaining as a result of pretreatment with radioactivity concentrations above certain levels are to be treated as radioactive wastes in the subsequent processes.

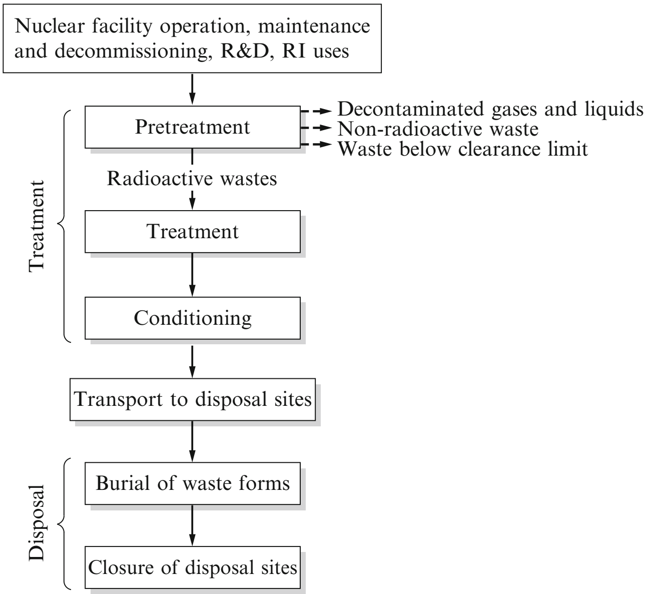


Fig. 1.2 Process flow from radioactive waste generation to disposal

Treatment is a process to improve safety and economy by changing the properties of radioactive wastes. It involves steps such as: filtration, ion exchange, and coagulation and sedimentation, to remove radionuclides; and incineration and compaction for volume reduction. Such parts of the wastes that are considered to have been decontaminated by the removal of radionuclides are no longer radioactive wastes.

Conditioning is a process to convert radioactive wastes into forms that are safe to handle, transport, store and dispose of. Wastes are solidified, sealed in containers, and additionally packaged to be transformed into waste forms. These steps may be easier to understand when called "waste form production." Conditioning is sometimes called packaging.

Waste forms are transported to disposal sites and emplaced in the disposal facility such as trench, vault or repository. In Japan today, all radioactive wastes are disposed of in the ground. Sea dumping, which used to take place overseas, has been banned, and other disposal methods once internationally discussed, such as sub-seabed disposal, ice sheet disposal and extraterrestrial disposal, are seldom mentioned today because of their lack of feasibility.

At a disposal site, when a specified volume of waste forms has been emplaced, that section of the site is backfilled; this act is called burial. When all burial sections are backfilled, the disposal site is closed. Closure is a physical step and usually does not mean the completion of the disposal project. After closure, institutional control including monitoring continues for a specified period of time. The form of institutional control varies depending on the type of disposal site. In geological disposal, there may be cases where waste forms remain ready to be taken out even after emplacement underground. These disposal concepts are discussed in Chap. 6, "Radioactive Waste Disposal."

Figure 1.2 illustrates the basic processes of handling radioactive wastes. In practice, some processes can vary between waste types. For example, some wastes are temporarily stored before proceeding to the next step. In particular, vitrified wastes, which are high-level radioactive waste resulting from reprocessing, are stored for 30–50 years for cooling. This kind of storage used to be called interim storage, but the term now refers to the step during which spent fuel waits to be reprocessed. In Japan, the place for interim storage is the Recyclable-Fuel Storage Center in Mutsu City, Aomori.

Although the above paragraphs mentioned that disposal means underground disposal in Japan, it should be noted that there are conceptually two fundamental principles of disposal: "dispersion and dilution" and "concentration and confinement." The latter refers to disposal aimed at minimizing environmental releases by reducing waste volume and making waste easier to manage, as in underground disposal. The former includes releasing gases and liquids to the atmosphere or the sea. The gases and liquids being released have radioactivity concentrations below specified limits and are not considered radioactive wastes by definition. In other words, they are outside the scope of regulation. This is why release to the atmosphere and the sea does not appear as processes after treatment in Fig. 1.2.

1.3.2 Systematic Management of Radioactive Wastes

The first priority in managing radioactive wastes is ultimately disposing of them safely, regardless of their characteristics and volume. Toward this goal, measures have been taken to safely dispose of all types of radioactive wastes listed in Table 1.2, while efforts have been made to restrict radioactive waste generation, such as the clearance system. These are for the purposes of reducing the risk source and curbing the cost of waste treatment and disposal.

The disposal depth for radioactive wastes varies by the waste type, but the choice of disposal depth—landfill disposal and concrete vault disposal at a shallow depth, subsurface disposal at a medium depth, or geological disposal deep underground—creates an order-of-magnitude difference in disposal costs. This suggests the possibility that if the radioactivity of waste to which subsurface disposal applies is reduced by decontamination to a level to which shallower concrete vault disposal is applicable, disposal costs may be curbed. By contrast, volume reduction, which is concentration process, increases radioactivity concentration and can result in the need for disposal at a greater depth. Treatments such as volume reduction, decontamination and solidification are inseparable from this kind of tradeoff concept.

As already mentioned, reprocessing has advantages and disadvantages of its own in waste management. Reprocessing is naturally effective in restricting high-level radioactive waste generation. Both the volume and total amount of high-level radioactive wastes for disposal are smaller when spent fuel is disposed of in the form of vitrified wastes than when it is directly disposed of. However, the benefit of using reprocessing in radioactive waste management will be limited if the total amount of wastes subject to geological disposal, including the geological disposal amount of TRU wastes generated from the operation of the reprocessing plant, is greater than the amount of spent fuel to be disposed of.

Partitioning and transmutation technology can probably reduce the potential hazards of high-level radioactive wastes or the duration of such hazards (Chap. 2, "Generation and Characteristics of Radioactive Wastes"). This technology may contribute to reducing the volume of waste forms and extending the service life of disposal sites. However, the technology will lead to the creation of wastes unexpected in the current nuclear fuel cycle, such as high-level radioactive wastes in different forms, secondary wastes, and wastes from the dismantling of related facilities. What is needed as motivation to drive the introduction of this technology is a quantitative prediction that waste management based on this technology will still be less burdensome than the current waste management, even with the treatment and disposal of such new types of radioactive wastes and necessary safety measures taken into consideration.

It would be ideal if radioactive wastes generated could be treated and disposed of under an integrated system that provides for safety and rationality for all kinds of radioactive wastes. Life cycle assessment may enable the establishment of such a system. Evaluation based on the life cycle assessment approach was once applied to the cost of nuclear energy, but it did not go far enough to reach the concept of radioactive waste management. For radioactive wastes, which are more traceable than industrial wastes because of the clearer flow of materials, there has been no comprehensive evaluation that covers every phase from generation to disposal. This is probably because no clear indicators (environmental impact indicators) like CO_2 emissions used for comparing different electric power sources have been established to measure the “impact” of radioactive wastes and the “burden” of radioactive waste disposal. How to establish the safest and most cost-effective radioactive waste management system and how engineering should contribute to it are always a challenge.

In summary, radioactive waste management encompasses policies such as the clearance system, which permits low-level radioactive wastes to be exempted from regulations, and disposal of high-level radioactive wastes, for which safety assessment may be required for the period of more than a few tens of thousands of years. These policies and regulatory systems reflect not only the characteristics of a country’s politics, including the distance between the public and government, but also that country’s ethics. The concept of securing safety in a remote future has never been discussed for conventional engineered artifacts, and may call for an innovative framework of thinking that connects the scientific and technological rationality and the public’s virtues and common sense. Technological efforts for radioactive waste management should offer quantitative and comprehensive basis for social decision-making.

References

1. International Atomic Energy Agency, *The Principles of Radioactive Waste Management*, IAEA Safety Series, No. 111-F (1995)
2. Nuclear Energy Agency of the Organisation for Economic Co-operation and Development, *The Environmental and Ethical Basis of Geological Disposal, A Collective Opinion of the NEA Radioactive Waste Management Committee* (1995)

Chapter 2
Generation and Characteristics of Radioactive Wastes

Minoru Okoshi and Shinichi Nakayama

2.1 Wastes from Uranium Mining and Melting Facilities

Mining refers to the act of collecting ore containing a target metal from mines. This process generates gangue—rock that is commercially valueless and therefore subject to disposal. The process of extracting metal from its ore is called melting. Uraninite, pitchblend and brannerite are uranium containing ores, and they contain uranium in the form of oxide. Their grade generally ranges from 0.1 to 0.3 % triuranium octoxide (U_3O_8) equivalent. Although extracted ore of ordinary metals undergoes “dressing,” the process of separating valueless rocks based on physical or chemical characteristics by such methods as fire refining and aqueous refining, the dressing process is not effective for low-grade uranium ore. This kind of ore is crushed into pieces and dissolved in acid or alkali solution. Then uranium is refined and concentrated, followed by precipitation using strong alkali. An intermediate from this milling process is uranium concentrate, which is U_3O_8 powder, called yellow cake for its color. Its uranium content (U_3O_8 content) is about 70–80 %. This material is further refined (or purified) to increase purity and is converted to forms such as UF_6, UO_2 or metallic uranium, suitable for use as reactor fuel in the next process at fuel fabrication facilities. In this connection, refining means increasing the purity of metal resulting from melting by electrolysis or other processes; in the uranium melting process, purification corresponds to refining.

Wastes generated from mining and melting processes mainly includes, in addition to gangue, refining effluent, tailings of ore after uranium extraction (also called

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The nuclear regulatory system in Japan has been changed significantly after the Fukushima Daiichi Nuclear Power Station accident in March 2011. Descriptions in this chapter have been translated from the book originally published in Japanese before the accident, with minimal update where appropriate.

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mill tailings), and wastes from facility maintenance. These wastes are managed under the Mine Safety Law and not classified as radioactive wastes in Japan. Melting effluent is neutralized and diluted before being released.

When a light water reactor with a rated electrical output of 1,000 MW is operating at an 80 % loading factor, 40,000 MW/d burn-up and 33 % thermal conversion rate, it consumes about 27 tons of low-enriched uranium fuel (4 %) annually. How much waste is generated from the mining and melting processes when this standard reactor is operated for 1 year can be estimated like this. For the sake of simplicity, it is assumed that uranium recovered by reprocessing is not recycled. Provided that the ^{235}U content in the depleted uranium generated by enrichment is 0.3 %, the amount of natural uranium needed for a 1 year period calculated from the mass balance in the enrichment process is approximately 244 tons (see Exercise 1 at the end of this chapter). Assuming that the uranium recovery rate during mining and melting is 93 % and the ore grade is 0.2 %, 130,000 tons of ore would have to be extracted and smelted.

Also included in the waste from mining and melting, besides residual uranium, is almost the entire amount of progeny nuclides derived from isotopes of uranium that decayed in the ore. These progeny nuclides are considered to be in the state of secular equilibrium with uranium isotopes while confined in ore. Once secular equilibrium is broken by melting, radiation thereafter attenuates in accordance with the half-life (about 80,000 years) of ^{230}Th, a progeny nuclide of ^{238}U. Among the progeny nuclides, volatile ^{222}Rn has a short half-life of 3.7 days, but it continues to be produced because of the decay of the parent nuclide. Special attention should be paid to ^{222}Rn in radiation control because it has high mobility in the environment. Waste generation from mining and melting is not a major issue in Japan, which has no large uranium mines. However, in Canada, one of the major uranium producing countries, tailings in storage have reached about 225 million tons, and their confinement and stabilization, as well as closure work at abandoned mines, are recognized as serious problems.

In veins in Kazakhstan that have attracted attention recently, where deposits are relatively low-grade and therefore mining and melting is not financially viable, “in-situ leaching” is used, which injects acidic solutions directly into the underground vein to dissolve uranium and draw up uranium solution out of downstream wells. Although this method does not generate waste as described above, the injected acid cannot be collected entirely. The operator must expect a certain level of groundwater contamination.

The collection of seawater uranium, a prospective method, is expected to allow the avoidance of uncollectable waste generation and environmental pollution problems.

2.2 Wastes from Fuel Fabrication Facilities

Fuel fabrication facilities are where nuclear fuel materials, such as uranium and plutonium, are physically or chemically treated to fabricate them into a form or a composition ready for use as reactor fuel. These are divided into two types: uranium processing facilities and mixed oxide (MOX) fuel fabrication facilities.

Table 2.1 Characteristics of wastes from fuel fabrication facilities [1]

Source facility	Conversion and enrichment facilities	Enrichment, reconversion and fabrication facilities	Enrichment facilities	MOX fuel fabrication facilities
Waste type	Uranium waste contaminated with natural uranium	Uranium waste contaminated with enriched uranium	Uranium waste contaminated with depleted uranium	TRU waste
Composition of isotopes of uranium	^{238}U: 99.2739 %	^{238}U: 95 % or more	^{238}U: 99.7 % or more	–
	^{235}U: 0.7204 %	^{235}U: 5 % or less	^{235}U: 0.3 % or less	
	^{234}U: 0.0057 %	^{234}U: around 0.1 %	^{234}U: around 0.005 %	
Coexisting nuclides	Uranium series nuclides, actinium series nuclides	Progeny nuclides (^{231}Th, ^{234}Th, ^{234}Pa)	Progeny nuclides (^{231}Th, ^{234}Th, ^{234}Pa)	Progeny nuclides (^{231}Th, ^{234}Th, ^{234}Pa), FPs

MOX mixed oxide, TRU transuranium, FP fission product

Uranium processing facilities consist of conversion facilities, which convert uranium oxide (U_3O_8) into uranium hexafluoride (UF_6); enrichment facilities, which enrich UF_6; reconversion facilities, which reconvert enriched UF_6 into enriched uranium oxide (UO_2); and fabrication facilities, which fabricate enriched uranium oxide into fuel assemblies. MOX fuel fabrication facilities are where uranium and plutonium which are recovered from reprocessing of spent fuels are converted and fabricated.

The operation of these facilities generates radioactive waste containing uranium or plutonium, or both. The generators of such wastes are limited to three: fuel fabricators, Japan Nuclear Fuel Ltd. (JNFL), and the Japan Atomic Energy Agency (JAEA), which operates enrichment facilities. Their wastes can be classified by the characteristics of contained radioactive material as shown in Table 2.1 [1]. The components contained in uranium wastes are essentially limited to uranium isotopes and their progeny nuclides, as far as radionuclides are concerned, although they can be natural uranium, enriched uranium or depleted uranium depending on the type of facility. And they contain no radionuclides produced by activation or nuclear fission. It should be noted that the uranium wastes from enrichment facilities in this table include those contaminated with depleted uranium, but depleted uranium itself is not considered as waste because it is a resource for future use as fast reactor fuel.

Wastes can be mainly classified into the following types according to their properties: used off-gas filters from gaseous waste treatment, sludge as a result of liquid waste treatment, combustible solid waste (e.g., used work clothes and gloves and waste wood), and used centrifugal separators. The amount of generation of these wastes is shown in Table 2.2 [1]. It is safe to say that the amount of uranium wastes generated is almost in proportion to the amount of fuel fabricated. In Japan,

Table 2.2 Uranium wastes generated per unit amount of uranium fuel fabrication (in the number of 200-liter drums per ton of uranium) [1]

Combustible waste	Incineration ash	Sludge	Incombustible miscellaneous solid wastes	Filters	Total (excluding combustible waste)
(1.36)	0.05	0.24	1.00	0.49	1.78

by the end of FY 2050, the cumulative amount of these wastes is estimated to reach approximately 115,000 tons (about 500,000 200-liter drums), including an estimated amount of clearance wastes of approximately 90,000 tons [2]. Table 2.3 indicates the average uranium content in different types of uranium wastes. The amount of incineration ash generated is small as shown in Table 2.2, whereas its uranium content is high as shown in Table 2.3.

Uranium-containing radioactive wastes from fabrication facilities are characterized as follows in comparison with other types of radioactive wastes [3].

1. The radionuclide content is relatively low (ranging from 10^4 Bq/g to 1 Bq/g, which is close to the natural content), and the contents of beta- and gamma-emitting nuclides are especially low.
2. Since uranium isotopes, which are primary radionuclides, have very long half-lives, the radionuclide content in wastes does not essentially decline. The radioactivity concentration will gradually increase with the generation of progeny nuclides and reach its peak in a few hundreds of thousands of years.
3. The gaseous progeny nuclide, ^{222}Rn, is generated, incurring the risk of a significant radiation dose depending on the condition.
4. The possibility that there is an effect from non-radiological factors (e.g., properties of uranium as a heavy metal) must be taken into consideration.
5. Consideration should be given to criticality control.

There is no special treatment method for uranium wastes that focuses on its radiological characteristics; uranium wastes are treated in the same methods as those used at nuclear power plants, according to the physicochemical characteristics of each radioactive waste. Meanwhile, to effectively use uranium resources and reduce the environmental impact of burial disposal, there are various decontamination methods to remove and recover uranium from generated radioactive wastes. For example, the ultrasonic washing method and the wet blast method are used for flame-resistant wastes, and the electropolishing method is additionally applied to carbon and stainless steels, allowing decontamination even to the background level in some cases [2].

The clearance levels for the metal wastes from uranium handling facilities were calculated by the former Nuclear Safety Commission of Japan in 2009 [4] (see Sect. 4.6.1, Clearance for uranium-handling facilities). Building on the calculation results, legislation is now being considered for uranium clearance levels. Metals below the statutory clearance levels will not be considered as radioactive wastes. By contrast, metals contaminated beyond the clearance levels will be treated as radioactive wastes and be disposed of by the appropriate method chosen from the existing disposal concepts according to their uranium content.

Table 2.3 Uranium content and amount in wastes from fuel fabrication [1]

	Combustible wastes	Incineration ash	Sludge	Incombustible miscellaneous solid wastes			Filters	Aggregate average
				Flame-resistant	Incombustible	Total		
Uranium content (g-U/kg)	4.2	41	8.0	2.1	0.6	1.0	16	5.5
Uranium amount (g-U/200-liter drum)	290	3836	806	138	43	86	481	460

2.3 Wastes from Nuclear Power Plants [5]

Nuclear power plants generate all forms of wastes—gaseous, liquid, and solid wastes. Their characteristics vary depending on the reactor type. The following sections describe the waste generated by boiling water reactors (BWRs) and pressurized water reactors (PWRs).

2.3.1 *Gaseous Wastes*

Gaseous wastes that should be considered at nuclear power plants are gaseous or volatile fission products. The radionuclides include ^{85m}Kr (half-life: 4.4 h), ^{87}Kr (78 min), ^{88}Kr (2.77 h), ^{133}Xe (5.27 d), ^{133m}Xe (2.3 d), ^{135}Xe (9.13 h) and ^{138}Xe (17 min), which are all radioactive noble gases, and ^{131}I (8.02 d), which is a halogen. Radioactive noble gases often have a relatively short half-life.

The gaseous waste treatment system of a nuclear power plant mainly consists of three subsystems: the reactor off-gas system, the standby gas treatment system, and the building air conditioning and ventilation system.

The filters used in the standby gas treatment system are designed to provide sufficient efficiency to remove 99.9 % or more of the iodine and particulate matter from the air released from the reactor building to the stack in the event of an accident. In a BWR, off-gas from the reactor is transferred to the turbine together with steam through the main steam pipe. The steam is condensed to water in the main condenser, while the off-gas is drawn with the air ejector, transferred to the gaseous waste system, goes through the decay pipe and the activated carbon type noble gas hold-up system for sufficient radioactive decay, before being released from the stack. In the event of a leak from a pump, valve or other components, the ventilation air in the reactor building or the turbine building may contain a trace amount of radioactive material. This air is released from the stack through the filter in the air conditioning and ventilation system.

A PWR has a closed circuit reactor cooling system, and it does not continuously release off-gas like the BWR. The amount of off-gas from the PWR is not as large as that from the BWR, and it is stored in the gas decay tank for a few tens of days for the decay of nuclides with short half-lives. In the PWR, hydrogen is added to the reactor coolant as a corrosion inhibitor. To keep its content at the proper level, hydrogen gas is injected in the volume control tank to maintain the specified pressure. At the startup and shutdown of the plant, hydrogen gas is replaced by nitrogen gas in the volume control tank. During this gas replacement process, noble gas in the primary coolant is discharged out of the system, along with hydrogen gas and nitrogen gas. To reduce the radioactivity of this noble gas, hydrogen gas in the off-gas is separated by the hydrogen recombiner first, and the remaining off-gas containing noble gas is stored in the gas decay tank for one to one and a half months for radioactive decay before being released to the environment. The latest PWR

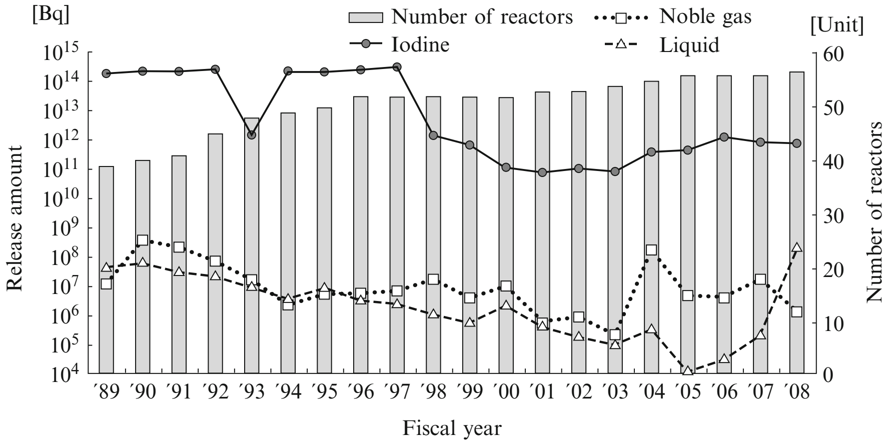

Fig. 2.1 Amount of radioactive gaseous and liquid wastes released from commercial power reactors and the number of reactors in Japan [6]

model uses the activated carbon type noble gas hold-up system, which requires simpler equipment than the volume control tank.

Figure 2.1 shows the trends in the total amount of gaseous wastes released from reactors in Japan. Since the beginning of this century, the released amount has been smaller by three to four orders of magnitude than in the 1990s. This is attributable to the shutdown of the Tokai Power Station of the Japan Atomic Power Company (JAPC). The reactor at the Tokai Power Station was a gas-cooled reactor, which, unlike light water reactors, directly released air used for cooling around the core to the environment and therefore released air containing ^{41}Ar (110 min).

2.3.2 Liquid Wastes [7, 8]

The liquid waste treatment system separates both radioactive effluent and effluent potentially contaminated with radioactive material generated in the power plant according to characteristics, then collects and treats them. The treated water is reused in the power plant or discharged to the environment after assessment of the radioactivity concentration. The total amount of liquid waste released from commercial reactors in Japan to the environment is shown in Fig. 2.1. The amount of liquid waste has increased since 2007 because of the contribution of JAPC's Tokai Daini Power Station. In the volume reduction process for miscellaneous solid wastes employed by this facility, radionuclides migrate to the off-gas where they become trapped in the washing water of the off-gas scrubber before being released as effluent.

Table 2.4 Example design specifications for the BWR condensate demineralizer using ion exchange resin

Condensate flow rate [m^3/h]	6,345
Number of demineralizer tanks	10 (nine demineralizers in operation, one on standby)
Volume of cation exchange resin [m^3/tank]	4.25
Volume of anion exchange resin [m^3/tank]	2.65
Total resin volume[a] [m^3]	70.5

[a] Including the volume of intermediate resin for chemical regeneration

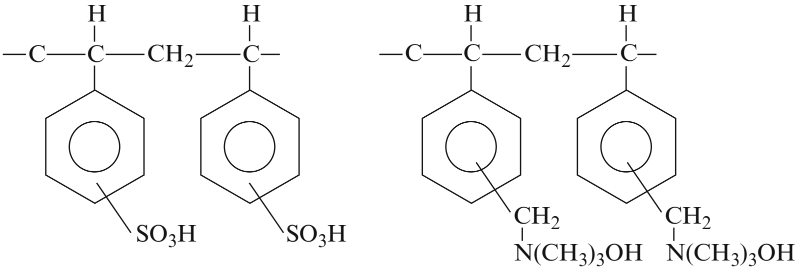

Fig. 2.2 Chemical structure of two ion exchange resins: *left*, cation exchange resin; *right*, anion exchange resin

In nuclear power plants, granular and powdery ion exchange resins are widely used in equipment such as the condensate demineralizer, condensate filter demineralizer system, reactor water purification system and effluent treatment system. Table 2.4 shows example design specifications for the BWR condensate demineralizer using ion exchange resin. The volume of ion exchange resin used for liquid waste treatment is as large as about 70 m^3. The chemical structure of an example ion exchange resin is shown in Fig. 2.2. It is based on a copolymer of styrene and divinylbenzene to which a sulfone group or a quaternary ammonium group has been introduced. Each group is capable of exchanging positive and negative ions. Radioactive material in liquid is fixed onto the ion exchange resin, which will then be treated as solid waste.

Liquid wastes are subject to filtration through which the solid component of effluent is separated and removed. The filter primarily employed for filtration in PWRs is the “hollow fiber” filter (HFF), which uses a hollow, filamentous membrane with microscopic pores on the surface. The HFF can be downsized by bundling filamentous filters while being able to minimize pressure losses arising from the passage of water. It also delivers such benefits as not easily clogging due to the small pore size, high performance and long life, thus reducing secondary waste generation. These treatment technologies are elaborated on in Chap. 5, “Radioactive Waste Treatment Technologies.”

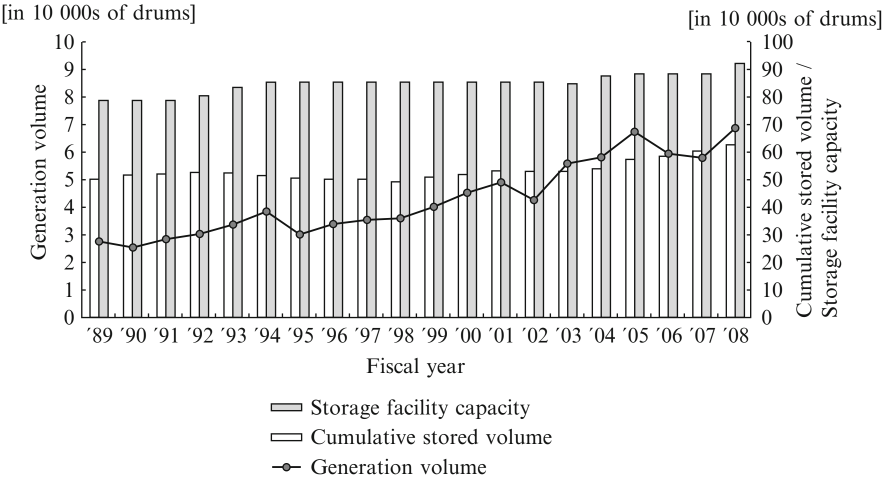

Fig. 2.3 Generation volume and cumulative stored volume of radioactive solid wastes from commercial power reactors in Japan [6]

2.3.3 *Solid Wastes*

Solid wastes from nuclear power plants are classified into the following types and managed according to their properties.

1. Sludge: This is stored in the sedimentation tank in the water purification system or solidified with cement or other material after storage for radioactive decay.
2. Spent resin: This is stored in the spent resin tank for radioactive decay, before being incinerated at miscellaneous solid waste treatment facilities or solidified with cement or other material. The annual generation volume of spent exchange resin per reactor is 20 m^3 for a BWR and 4–7 m^3 for a PWR.
3. Combustible miscellaneous solid wastes: These are incinerated in incineration facilities for volume reduction. Incineration ash is stored in metal drums.
4. Incombustible miscellaneous solid wastes: These are compacted for volume reduction if possible. In some power plants they may subjected to melting.
5. Relatively high level radioactive solid wastes: Activated components, such as spent control rods, have a relatively high level of radioactivity and are therefore stored in the fuel pool for radioactive decay. For effective storage of fuel in the fuel pool, some power plants have a side bunker facility as a storage pool dedicated to receiving wastes.
6. Solid wastes are usually stored in 200-liter drums or metal containers of around 1 m^3, which are called square containers.

Figure 2.3 shows example data regarding the generation of solid wastes from the Japanese plants. In FY 2008, 734 m^3 of waste was generated in total from three power plants due to the replacement of steam generators, reactor vessel upper heads and reactor core internals. The graph indicates a gradual increase in the annual waste generation volume because of the increase in the number of operating plants and the extension of periodic inspection periods as plants age.

In FY 2008, commercial power reactors in Japan generated approximately 67,600 200-liter drums of low-level radioactive wastes. However, the cumulative stored volume increased only about 21,700 drums. This is attributable to activities other than waste volume reduction, such as shipment to the Low-Level Radioactive Waste Disposal Center of JNFL, located in Rokkasho Village, Aomori Prefecture, and clearance by JAPC, along with volume reduction through incineration and compaction. The cumulative stored volume in FY 2008 was about 624,300 drums, which accounts for 68.3 % of the capacity of solid waste storage facilities, 914,600 drums.

The data in Fig. 2.3 shows statistics on commercial power reactors only, excluding waste from Fugen and Monju, which are R&D phase reactors at JAEA. Fugen, which is now being decommissioned, generated about 160 200-liter drums of low-level radioactive waste in FY 2009; its cumulative stored volume has reached approximately 19,100 drums, representing 88 % of the storage capacity [9]. Ion exchange resin and filter sludge are stored in the tanks, and spent control rods and neutron detectors in the spent fuel pool. Monju, which resumed operation in May 2010 after an interval of 14 years, generated 584 drums of waste in FY 2009 even while out of service; its cumulative stored volume is 4,432 drums [9].

Solid wastes generated as a result of reactor operation and carried out of power plants are subject to landfill disposal, concrete vault disposal or subsurface disposal according to the radioactivity level (Chaps. 1 and 6).

2.4 Wastes from Reprocessing

Radioactive wastes from reprocessing plants are characterized by a greater content of TRU nuclides than other types of radioactive wastes. This section describes radioactive wastes from reprocessing plants using the tried-and-tested PUREX (Plutonium and Uranium Recovery by EXtraction) method. Spent fuel (including cladding and other components of fuel assemblies), which is the subject of reprocessing, is highly radioactive, containing approximately 31.6 PBq ($=31.6 \times 10^{15}$ Bq) of radioactivity per ton of waste, as exemplified in Table 2.5 [10].

The PUREX method separates uranium and plutonium, as nuclear material, from fission products by dissolving spent fuel with nitric acid into a solution, which then repeatedly undergoes the same extraction process. Since it is an aqueous method, large amounts of liquid wastes are produced; the liquid wastes contain most of the fission products that were in the spent fuel, unrecovered uranium and TRU nuclides.

Figure 2.4 shows radioactive wastes generated in different stages of reprocessing based on the PUREX method. Radioactive liquid wastes mainly consist of high-level effluent and acid effluent from solvent extraction process. The high-level effluent is vitrified as high level solid radioactive waste. Various kinds of chemicals are used in reprocessing, such as nitric acid to dissolve the fuel, tributyl

Table 2.5 Data on spent fuel [10]

Fuel	Weight	Uranium	957 kg
		Plutonium	9 kg
		Fission products	34 kg
	Radioactivity	Alpha- emitting nuclides	4.0 PBq
		Beta- and gamma-emitting nuclides	27.4 PBq
	Calorific power		2.96 kW
Fuel assembly components	Weight	Zircaloy	292 kg
		Stainless steel	106 kg
		Inconel	24 kg
		Alumina	2 kg
	Radioactivity	Activated nuclides	0.2 PBq*

Data are per ton of new fuel uranium, for PWRs, at a burn-up of 33,000 MWd, and for fuel having been cooled for 3 years after being unloaded from the reactor (except were marked with an asterisk [*], which is for fuel cooled for 10 years after being unloaded from the reactor)

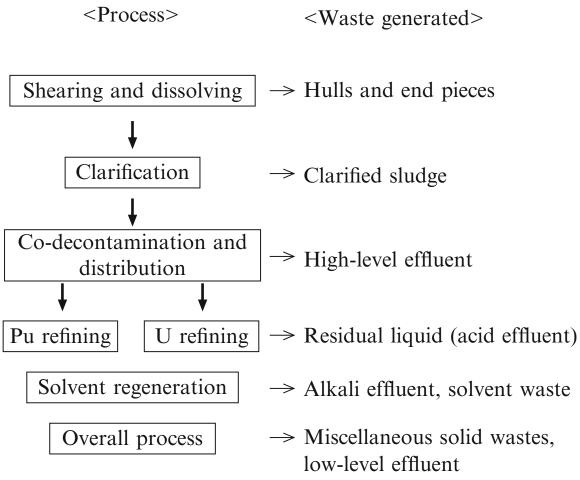

Fig. 2.4 Major radioactive wastes from reprocessing based on the PUREX method [11]

phosphate (TBP) to extract uranium and plutonium, dodecane as an organic solvent to dilute the TBP, and sodium carbonate as a cleaner for the organic solvent. Although some of these are regenerated and reused, all will ultimately become radioactive wastes in the form of spent solvents.

Solid wastes can be divided into several types: end pieces of fuel structural components generated during the shearing and dissolution processes, and hulls used as cladding, which are both wastes with a relatively high level of radioactivity; and combustible or incombustible low-level radioactive miscellaneous solids, which consist of waste components and materials from plants. In addition, repaired or replaced items from different stages of reprocessing, such as primary equipment, auxiliary equipment and purification equipment, are contaminated with radioactive materials and treated as radioactive wastes. Reprocessing plants are decommissioned after their service life, leading to the generation of dismantling waste.

The reprocessing plant of JNFL has the capacity to reprocess 800 tons of spent fuel annually and is expected to generate 5,600 m^3 of waste every year.

2.4.1 Gaseous Wastes

When spent fuel is sheared and dissolved, radioactive gas trapped in the fuel pin plenums or fuel pellets is released. This gas constitutes a large part of the radioactive gaseous wastes from reprocessing plants and its major nuclides are Kr and I.

More than 80 % (in molar ratio) of the gas generated as fission products in nuclear fuel is Xe. However, due to its short half-life (9.1 h for ^{131}Xe, 5.27 d for ^{133}Xe), most of it decays during the cooling period before reprocessing starts. Among the major nuclides of Kr and I in gaseous wastes, the dominant one is ^{85}Kr (10.3 y). Although its collection was considered for the purpose of reducing the environmental release of radioactivity, such a measure has not been adopted by JNFL's reprocessing plant. To collect and solidify Kr, highly enriched krypton gas would need to be accumulated in the plant. The above decision not to recover Kr was made because dilution and diffusion through release from the main stack is more effective in reducing radiation doses to residents around the plant. Likewise, Kr is not collected in reprocessing plants in France and the U.K. either, where commercial reprocessing takes place [12].

JNFL's reprocessing plant is estimated to generate 1.23×10^{12} Bq (approx. 189 kg) of ^{129}I (15.7 million years) annually. On the other hand, ^{131}I (8.02 d) and other radioactive iodine nuclides with short half-lives disappear for the most part during the cooling period before reprocessing, at least temporarily. However, a very small amount, or 1.86×10^{11} Bq (approx. 40 μg) annually, of ^{131}I is estimated to be generated due to nuclear fission in high level liquid waste tanks and other parts of the plant [13].

Iodine can be removed either by the washing method or the solid adsorption method. A common washing method is the alkali washing method, which uses sodium hydroxide. Solid adsorbents being considered or actually used are silver adsorbents such as silver zeolite, silver silica gel, and silver alumina. The alkali washing method usually brings off-gas, containing iodine, in contact with 1–2 mol/L of sodium hydroxide (NaOH) to remove iodine as sodium iodide (NaI) or sodium iodate ($NaIO_3$).

$$3I_2 + 6NaOH \rightarrow 5NaI + NaIO_3 + 3H_2O$$

Washing with alkali solution is effective for removing inorganic iodine but not for organic iodine, which is difficult to capture.

Impregnated charcoal, which is made by adding a small amount of potassium iodide to activated carbon, is used as the iodine filter at nuclear power plants, but not at reprocessing facilities because iodine may react with NO_x in the off-gas and create an explosive compound. Unlike silver adsorbents, activated carbon only physically adsorbs iodine to temporarily hold it. Although adsorption efficiency increases as temperature drops, a reduction in the adsorption amount or desorption of the adsorbed iodine occurs if temperature rises or if other materials, such as water, are adsorbed. Therefore, if humidity is high and an adsorptive gas (e.g., acid gas and organic gas) coexists, the iodine capture efficiency significantly drops [10].

2.4.2 Liquid Wastes

Acidic PUREX raffinate from co-decontamination and partition processes during reprocessing contains most of the nonvolatile fission products included in spent fuel, actinides, and trace amounts of uranium and plutonium (about 1 % of the amount in spent fuel), as well as other corrosion products, and is highly radioactive, at about 10^7 GBq/m^3. Reprocessing using the PUREX method produces about 5 m^3 of the raffinate from every ton of spent fuel. Its volume is reduced to about 1/10 through evaporation. The volume reduction rate is closely related to the burn-up and the cooling time of the fuel to be treated. If the cooling period is long enough, the limiting factor of the extent of volume reduction is the heat generation in the concentrated solution and the amount of solids in the liquid waste, which contain fission products and chemicals used for reprocessing. The appropriate volume reduction assumed for the design of the storage tank capacity at a reprocessing plant is a reduction to around 0.6 m^3 per ton of spent fuel. To prevent metallic corrosion of the evaporator, the raffinate undergoes treatment to lower the nitric acid concentration by reducing the nitric acid in the solution to NO_x by adding formaldehyde and sucrose. Then vitrification, described in the next section, takes place.

Liquid wastes from other reprocessing processes, such as uranium refining and plutonium refining, also undergo evaporation. In overseas reprocessing plants, where liquid wastes have high ^{90}Sr and ^{137}Cs contents, a coagulation-sedimentation treatment is used to coprecipitate ^{90}Sr with sulfate and ^{137}Cs with nickel ferrocyanide for removal. Concentrated liquid waste and sludge generated in the course of concentration of these liquid wastes are usually subjected to bituminization. Due to differences in radiation levels in liquid waste, bituminization uses straight asphalt at nuclear power plants and blown asphalt at reprocessing plants in general. Blown asphalt is characterized by smaller cubical expansion caused by gas generated by radiolysis than straight asphalt. At JNFL's reprocessing plant, concentrated liquid waste from the evaporation of low-level radioactive liquid waste is dried and compaction-molded before being stored in metal drums.

The solvent used in reprocessing contains impurities which have been generated by chemolysis or radiolysis such as degradation products of TBP and dodecane.

The degradation products of TBP and dodecane include dibutyl phosphate, butyl nitrate and 1-butanol, and lauric acid, respectively. Solvent waste as a result of purifying spent solvent was once treated by incineration. However, since it causes significant corrosion to the incinerator, other methods, including vacuum distillation method and the TBP extraction method using phosphoric acid, are used today. Solvent waste mainly consisting of TBP separated by any of these methods is thermally decomposed, and the remaining phosphoric acid is neutralized with calcium hydroxide to get an inorganic salt. At JNFL's reprocessing plant, solvent waste from the solvent treatment system in the solvent collection facility is thermally decomposed, compaction-molded, and stored in metal drums or other containers.

2.4.3 Solid Wastes

Solid wastes, such as cladding (hulls) and fuel assembly components which are insoluble in nitric acid, are contaminated with not only undissolved fuel and fission products but also activated products that are radioactive as a result of irradiation with neutrons. The zircaloy hulls for light water reactors occlude tritium. Although discussions have been held on volume reduction methods for these solid wastes, they are stored without being treated at present. Other solid wastes generated during the reprocessing process undergo volume reduction by incineration, compaction or other methods.

The primary wastes from reprocessing plants are so called "TRU waste" and high-level radioactive wastes, as mentioned above. The former includes spent silver adsorbent, which has solidified ^{129}I, TRU waste from MOX fuel fabrication facilities (Sect. 2.2), and repatriated low-level radioactive wastes that have been generated in reprocessing projects contracted to overseas facilities. Repatriated wastes are elaborated on in Sect. 2.6.

As explained in Chap. 1, TRU wastes from the reprocessing process are characterized as being contaminated with both nuclear fission products and TRU nuclides and as containing diverse waste forms; therefore they range widely in radioactivity concentration. In these aspects, TRU wastes are unlike wastes from fuel fabrication facilities and from nuclear power plants, which generate wastes of almost uniform composition. The radioactivity concentrations in TRU wastes (Fig. 1.1) from alpha-emitting nuclides are from 10 kBq/ton to 1 TBq/ton and from beta- and gamma-emitting nuclides, 1 MBq/ton to 1 PBq/ton. The appropriate disposal method is selected for each type of TRU waste in consideration of such radioactivity concentrations and confinement performance for the given waste form.

Another waste specific to reprocessing is high-level radioactive liquid waste. This waste is mixed with glass material and solidified in stainless steel canisters after which it is converted into vitrified waste. In Japan, the term high-level radioactive wastes always refers to vitrified wastes. Table 2.6 shows major radionuclides contained in vitrified wastes and their radioactivity. They can be broadly

Table 2.6 Major radionuclides contained in vitrified wastes and their radioactivity[a] [14]

	Fission products			Transuranium		
Half – life (year)	Nuclide	Half – life (year)	Radioactivity (Bq)	Nuclide	Half – life (year)	Radioactivity (Bq)
2–100	^{90}Sr -^{90}Y	29.1	6.41×10^{15}	^{238}Pu	87.8	3.39×10^{11}
	^{125}Sb -^{125m}Te	2.8	1.32×10^{14}	^{241}Pu	14.4	1.01×10^{13}
	^{134}Cs	2.1	1.74×10^{15}	^{243}Cm	28.5	8.95×10^{11}
	^{137}Cs -^{137m}Ba	30.2	9.32×10^{15}	^{244}Cm	18.1	1.67×10^{14}
	^{147}Pm	2.6	1.97×10^{15}			
	^{151}Sm	90.0	1.38×10^{13}			
	^{152}Eu	13.6	6.85×10^{10}			
	^{154}Eu	8.6	1.94×10^{14}			
	^{155}Eu	5.0	8.99×10^{13}			
100–1,000				^{241}Am	432	5.52×10^{13}
				^{242m}Am	152	2.86×10^{11}
1,000–10,000				^{240}Pu	6,540	4.51×10^{10}
				^{243}Am	7,380	1.26×10^{12}
				^{245}Cm	8,500	3.13×10^{10}
				^{246}Cm	4,730	5.81×10^{9}
10,000–1 million	^{79}Se	6.50×10^{4}	1.59×10^{10}	^{239}Pu	2.41×10^{4}	2.78×10^{10}
	^{99}Tc	2.13×10^{5}	6.59×10^{11}	^{242}Pu	3.87×10^{5}	2.18×10^{8}
	^{126}Sn	1.00×10^{5}	3.22×10^{10}			
1 million or longer	^{93}Zr	1.53×10^{6}	8.95×10^{10}	^{237} Np	2.14×10^{6}	1.32×10^{10}
	^{107}Pd	6.50×10^{6}	5.96×10^{9}			
	^{135}Cs	2.30×10^{6}	2.22×10^{10}			
Total radioactivity		2.28×10^{16} Bq				
Element weight		38.4 kg		Np 506 g, Pu 22.1 g, Am 583 g, Cm 61.2 g		

[a] The figures are per ton of spent fuel at a 45,000 MWd/ton burn-up in a PWR after a 5-year cooling period. The ratios transferred to wastes through reprocessing are assumed to be 0.2 % for U, 80 % for Np, and 0.2 % for Pu. Although ^{129}I (1.57×10^{7} years, 3.16×10^{7} Bq) contained in spent fuel is an important nuclide, it is not contained in vitrified wastes

divided into three types: fission products of uranium and its fissile products such as plutonium, TRU which are generated when uranium absorbs neutrons, and activation products which are produced from small amounts of impurities in fuel. Radioactivity levels of vitrified waste at the time of fabrication is dominated by nuclides with relatively short half-lives, such as ^{90}Sr (29.1 y), ^{137}Cs (30.2 y) and ^{244}Cm (18.1 y), with a total radioactivity of about 1×10^{16} Bq/ton. After these nuclides decay in a few hundreds of years, radioactivity remains from nuclides with long half-lives, such as ^{99}Tc (213,000 y), ^{93}Zr (1.53 million years) and ^{237}Np (2.14 million years), although they are present in small amounts.

When spent fuel is reprocessed within a few years after its unload from the reactor core, the decay of ^{241}Pu (14.4 y) is negligible. However, if reprocessing does not take place for 10 years or more after unload from the reactor core, generation of ^{241}Am from the beta decay of ^{241}Pu is not negligible. In its half-life of 432 years, ^{241}Am undergoes alpha decay to produce ^{237}Np. Reprocessing of spent fuels at an early point after unload and early Pu recycling after reprocessing are preferable because, from the viewpoint of Pu resource utilization, useful ^{241}Pu which has the large cross section of fission decreases by its decay and, from the viewpoint of the safety of geological disposal, the heat generated from ^{241}Am remains dominant for more than 1,000 years, and long-lived, toxic ^{237}Np is generated.

Vitrified wastes produced after reprocessing have very high levels of radioactivity and dose equivalent rates at the time of production are estimated as 14,000 Sv/h on the waste surface and 420 Sv/h at 1 m apart from the surface. Therefore, verification of canister integrity is performed remotely as a matter of fact. Because of high calorific values, vitrified wastes are cooled for 30–50 years after vitrification, followed by transportation to a geological disposal site. The cumulative generation amount of vitrified wastes is estimated to reach about 40,000 canisters, or about 7,000 m^3, by 2020 in Japan.

Vitrified wastes have a very long "life" because of the long-lived nuclides contained in the high-level radioactive wastes. This is the fundamental reason why geological disposal, which requires no further direct control after closure, is chosen. Figure 2.5 illustrates changes in radioactivity level in nuclear fuel throughout its life cycle, from uranium ore to power generation and disposal. To supply 1 ton of uranium for fuel, 750 tons of 1 % grade uranium ore is needed, which has a radioactivity of 1,000 GBq (1 % is a fairly high grade, and Canada has ore deposits with a uranium grade of this level). When fabricated into fuel and loaded in the reactor, radioactivity per ton of fuel drops to 100 GBq, but once fuel is used for power generation and is irradiated in the reactor, its radioactivity steeply rises by eight orders of magnitude, or 100 million-fold. During the 4-year period between unload from the reactor and reprocessing and vitrification, radionuclides with very short half-lives decay and disappear, reducing radioactivity by about two orders of magnitude. This is considered the radioactivity level for vitrified waste. Its radioactivity slowly declines by about three orders of magnitude for the first few hundred years because of the decay of radionuclides that played a dominant role in the initial radioactivity, such as ^{90}Sr, ^{137}Cs and ^{244}Cm. Although the radioactivity level

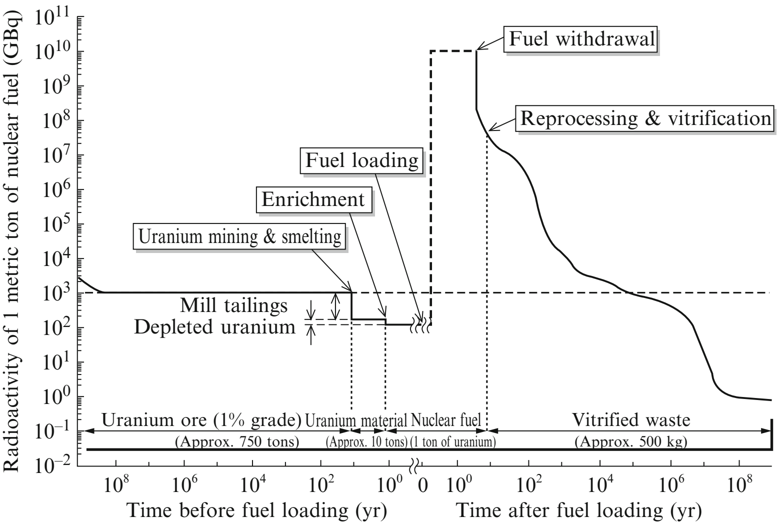

Fig. 2.5 Changes in radioactivity per ton of uranium throughout the nuclear fuel cycle (for 1 metric ton of 4.5 % enriched nuclear fuel) [15]

continues to drop thereafter as well, it takes about 100,000 years after terminating power generation until the level equivalent to the original uranium ore is restored.

Figure 2.5 also indicates that specific radioactivity 100 million or more years ago was higher than it is now. This is because the isotopic ratio of ^{235}U, with a 700 million year half-life, was greater at the birth of the Earth. It is easily calculated that the isotopic ratio of ^{235}U about two billion years ago was a few percent, far higher than it is now, 0.72 %. At that time, unevenly distributed uranium in underground veins and surrounded with groundwater and rocks (mainly silica) that were effective neutron moderators, created a "natural reactor." It has been confirmed that in the Oklo mine in the Gabonese Republic, Africa, a chain reaction of nuclear fission had occurred intermittently for a few hundreds of thousands of years [16].

2.5 Wastes from R&D and Radioisotope Uses

Radioisotopes (RIs), radiation generating devices, and nuclear fuel material are used not just in nuclear power generation and other energy fields but also for many other purposes, including R&D, agriculture, industry and health care.

RIs and radiation generating devices are used in industrial fields such as paper-making, steel sheet manufacturing (to make thickness measurements) and semiconductor fabricating, and in medical fields such as nuclear medical diagnosis (e.g., positron emission tomography-computed tomography, PET-CT) and the sterilization of injection needles and other medical equipment. Universities and research institutes conduct studies by using RIs as tracers and employ accelerators as radiation generating devices. The operation of these RI-using facilities and radiation generating devices generates RI-containing wastes such as test tubes, injectors, paper towels and off-gas filters. The dismantling of these facilities also generates such wastes as metals and concrete contaminated with RIs and activated products of these materials. These are collectively called RI wastes.

Typical radionuclides contained in RI wastes are ^{3}H, ^{14}C, ^{32}P and ^{35}S in the R&D and industrial fields and ^{99m}Tc, ^{125}I and ^{201}Tl in the medical field. Most RI wastes generated in Japan are collected and stored by the Japan Radioisotope Association (JRIA).

R&D activities at universities and research institutes produce a wide variety of wastes contaminated with nuclear fuel materials. Likewise, the dismantling of facilities handling nuclear fuel materials generates contaminated metals and concrete as well as activated reactor core internals. These are classified as radioactive wastes generated from research, industrial and medical facilities. Wastes from JAEA's prototype advanced thermal reactor Fugen and the prototype fast breeder reactor Monju are included in the wastes because they are in the R&D phase and not commercial reactors (Sect. 2.3.3). Typical radionuclides in the radioactive wastes are ^{3}H, ^{14}C, ^{60}Co, ^{90}Sr, ^{137}Cs and ^{238}U. Most of these wastes are stored at the business site of the source facilities.

RI wastes and radioactive wastes from laboratories are together called radioactive wastes from research facilities. These two types of wastes are often discussed separately because each is governed by a different set of regulations in Japan. The use of RIs is regulated by the Law Concerning Prevention from Radiation Hazards due to Radioisotopes, etc. (Radiation Hazards Prevention Law), the Medical Service Act, the Pharmaceutical Affairs Act, the Veterinary Practice Act, and the Clinical Laboratory Technician Act. The Radiation Hazards Prevention Law applies to about 4,600 business sites, of which about 1,000 are generators of RI wastes. The Medical Service Act, the Pharmaceutical Affairs Act, and the Clinical Laboratory Technician Act apply to about 1,300 facilities handling radioactive drugs. The use of nuclear fuel materials is governed by the Act on the Regulation of Nuclear Source Material, Nuclear Fuel Material and Reactors. Radioactive wastes from laboratories are generated from about 170 organizations, including JAEA, universities, and private enterprises.

It was decided in 2008 that radioactive wastes from research facilities should be disposed of by JAEA, and the Japan Atomic Energy Agency Act was revised accordingly. Survey results on the generated amounts of such wastes expected to be disposed of by FY 2048 are shown in Table 2.7 [17]. One notable characteristic of radioactive wastes from research facilities is the variation in radioactivity concentration in the waste forms, which ranges from somewhere around clearance levels to above the upper concentration limits for concrete vault disposal, because of the wide variety of sources. The disposal method appropriate for the radioactivity concentration of each waste must be selected accordingly.

Table 2.7 Estimated disposal amount of radioactive wastes research facilities [17]

Institution	Landfill disposal	Concrete vault disposal	Subsurface disposal
JAEA	244,400	188,700	56,200
JRIA	33,300	9,100	–
Universities, businesses, etc.	42,800	2,300	4,400
Total	320,500	200,100	60,600

Data are expressed in the number of 200-liter drums and indicate cumulative totals expected by FY 2048. The figures are not generation amounts at facilities but the number of disposal waste forms that will have undergone incineration, compaction or other treatment after generation

2.6 Repatriated Wastes

As a transitional measure until the inauguration of JNFL's reprocessing plant in Rokkasho Village, Aomori, Japanese electric utilities have outsourced the reprocessing of their spent fuel to AREVA NC (formerly COGEMA), based in France, and British Nuclear Group Sellafield Ltd. (BNGS, formerly BNFL). Approximately 7,100 tons of spent fuel has been sent overseas reprocessing by 2007. The recovered uranium and plutonium are repatriated from the reprocessing plants as fuel. Wastes generated during reprocessing are also returned to Japanese utilities because both European countries follow the policy of repatriating radioactive wastes from reprocessing to the country of origin, in principle. Such wastes are called repatriated waste.

Vitrified high-level radioactive wastes have been shipped to Japan 13 times by March 2010, with 1,338 canisters accepted in total since the first shipment of 28 canisters in April 1995 from the Port of Cherbourg, France, to Mutsu-Ogawara Port in Aomori. The ship used is owned by PNTL, an affiliated company of BNGS. After arrival at the port, waste forms are taken to JNFL's Vitrified Waste Storage Center in Rokkasho Village by a special land carrier. Approximately 850 canisters of vitrified high-level radioactive wastes are expected to be repatriated from the U.K. The Vitrified Waste Storage Center's storage facility employs a multi-stack vault storage structure with an air cooling system and has a storage capacity of 1,440 canisters, which is expected to increase to 2,880 in the future.

Low-level radioactive wastes are expected to be duly repatriated as well. France and the U.K. proposed changes in the specifications of repatriated wastes regarding this. The former proposed that the form of the TRU wastes (low-level radioactive wastes subject to geological disposal) be changed from the originally planned bituminized form (approx. 1,100 drums) to 28 canisters of vitrified waste. The U.K. proposed that a certain numbers of low-level radioactive waste forms—namely, about 4,500 drums of cemented TRU wastes subject to geological disposal and about 6,000 drums of heterogeneous solid wastes subject to shallow underground disposal—be exchanged with about 150 canisters of vitrified high-level radioactive wastes for repatriation. These proposals are expected to benefit Japan in that they will significantly cut back transportation costs because of the reduced volume of the waste to be repatriated and thus a smaller number of shipments will be required; as well they will create an extra allowance in Japan's storage capacity.

However, before these proposals could be accepted, the Japanese government had to revise laws concerning geological disposal. The British proposal on TRU wastes called for "exchanging with small amounts of high-level radioactive wastes (originating in overseas reactors) with equivalent radiation" (equivalent exchange). The then-applicable Designated Radioactive Waste Final Disposal Act [18], enforced in 2000, assumed accepting only high-level radioactive wastes generated at domestic power reactors and not those from overseas reactors. In 2005, the Act was amended to add TRU wastes to the list of the specified radioactive wastes and to oblige JNFL and JAEA, as the generators of TRU wastes, as well as electric utilities as the importers of high-level radioactive wastes to be received in place of TRU wastes, to contribute toward disposal expenses. This has legally enabled the acceptance of repatriated wastes.

2.7 Wastes Below the Clearance Limit

Clearance is discussed in further detail in Chap. 4. The clearance level is a radioactivity concentration for which radiation effects on the human body are small enough to be negligible, and materials with a radioactivity concentration below this level do not have to be classified as radioactive materials. Wastes with radioactivity below the clearance limit are outside the scope of regulation and are no longer considered as radioactive wastes.

Some of the wastes generated during nuclear facility dismantling are clearance items. When a 1,100 MW light water reactor is dismantled, approximately 500,000 tons of waste is generated, of which cleared items account for 2–5 %.

Meanwhile, there is the concept of "non-radioactive wastes." These are a kind of waste that is generated within a controlled area but they are distinguished from radioactive wastes because of usage records indicating no possibility of contamination with radioactive materials or no need for consideration of the impact of neutron-induced activation. Non-radioactive wastes account for 92–96 % of the wastes from the dismantling of a 1,100 MW light water reactor. This means that as far as the reactor is concerned, wastes below the clearance limit almost always refer to non-radioactive wastes.

2.8 Wastes from Partitioning and Transmutation

Partitioning and transmutation (P&T) technology separates long-lived radionuclides contained in high-level radioactive wastes and converts them into shorter-lived or stable nuclides through transmutation. Its primary goal is to reduce the long-term burden of geological disposal by attenuating the radioactivity of the high-level radioactive wastes in shorter time periods. The partitioning of elements contained in wastes is sometimes conducted, even without transmutation, for the purpose of optimizing treatment and disposal by developing the optimal waste form for each element, as in the case of the Hanford Site in the U.S., where high level effluent from nuclear weapons production is treated. However, the mainstream trend in the world is R&D focusing on systems combining both P&T, since transmutation has the potential to change long-lived nuclides into shorter-lived nuclides and partitioning is indispensable to transmutation.

How the introduction of this technology could change the properties of high-level radioactive waste, that is to say, how it could change radiation attenuation, has been well reported for many years [19]. However, introducing P&T technology means changing the amounts and flows of materials in the nuclear fuel cycle, which would result in the generation of radioactive wastes that are different in composition, radioactivity and chemical characteristics than wastes generated in the current nuclear fuel cycle. Just as all types of current radioactive wastes are ensured safe disposal in accordance with their characteristics on the basis of comprehensive discussions, all the radioactive wastes from the nuclear fuel cycle involving P&T call for consistent waste management policies. Furthermore, it is necessary to present benefits large enough to offset the economic and social cost of introducing the additional treatment of P&T.

This section first describes P&T technology and then analyzes and discusses radioactive wastes that would be generated if this technology were to be introduced. Medical and industrial applications of radionuclides separated from high-level radioactive liquid waste, or so-called effective utilization of waste, have been studied for a long time. Reference [20] is a starting point to provide further information.

2.8.1 Partitioning

This is a process to separate components subject to transmutation from spent fuel and high-level radioactive liquid waste generated from spent fuel reprocessing. The nuclides it mainly deals with are such actinide elements as ^{237}Np (2.14 million years), ^{243}Am (7,380 y) and ^{245}Cm (8,500 y) as well as such fission products as ^{99}Tc (213,000 y), ^{129}I (15.7 million years) and ^{135}Cs (2.3 million years). These are all long-lived radionuclides. Np, Am and Cm are called minor actinides because their weight content in spent fuel from light water reactors is smaller than that of Pu by more than one order of magnitude. Although such nuclides as ^{79}Se (65,000 y), ^{93}Zr (1.53 million years), ^{107}Pd (6.5 million years) and ^{126}Sn (approx. 100,000 y) merit attention from the viewpoint of the nuclide confinement capability of the geological disposal system, the above-mentioned ^{99}Tc, ^{129}I and ^{135}Cs are considered for partitioning because of their volume of production. Other important nuclides for partitioning are ^{90}Sr (29.1 y) and ^{137}Cs (30.2 y), which have relatively short half-lives and are therefore not subject to transmutation, but are limiting factors in disposal site design because of their calorific value.

Thus a wide variety of elements is considered for partitioning, and different partitioning technologies would be used depending on the type of waste. Aqueous partitioning technology would be employed for separation from high-level radioactive liquid waste, while an improved reprocessing technology based on the PUREX method would primarily be used for direct separation from spent fuel. Since transmutation efficiency is generally low, the transmutation process must be repeated. This means spent fuel that has undergone transmutation would be reprocessed. Since it would contain a drastically increased content of minor actinides, consideration on material degradation due to irradiation (e.g., damage to solvent) and criticality control measures would be needed. Because of solvent degradation, dry partitioning technology not using organic solvent might be a better candidate.

2.8.2 Transmutation

The transmutation method is essentially limited to one that uses neutrons. In other words, the primary nuclear reaction is nuclear fission in minor actinide transmutation, and neutron absorption in fission product transmutation. Since multiple nuclear reactions take place simultaneously in the reactor, the number of neutrons involved in nuclear fission reactions is important to increase the efficiency of the target transmutation. Fast neutrons are especially effective because minor actinides are more likely to cause nuclear fission than absorption in the 700 keV or higher neutron energy range. Therefore, transmutation requires a high-intensity, high-energy neutron source. The two most likely sources are fast neutrons in fast reactors and fusion reactors, and neutrons generated by nuclear spallation reactions in accelerators.

The method using fast reactors has the advantage of requiring fewer items to be developed because it is an extension of current reactor technology and nuclear fuel cycle technology. By contrast, the idea of using the accelerator involves many new R&D challenges, such as developing targets inducing the nuclear spallation reactions and the steady operation of the accelerator. However, the latter does not impose as difficult safety requirements on criticality as the former because the accelerator would maintain steady output in a subcritical state. A system that induces transmutation externally by an accelerator is called an accelerator-driven system (ADS).

2.8.3 Fuel for Transmutation

The chemical composition requirements for fuel used for transmutation are essentially the same as those for candidate materials for the current fast reactor fuel, such as oxides, metals and nitrides, although depending on the transmutation method to be used. The only difference is the content of minor actinides. To achieve high efficiency in transmutation, fuel must have greater minor actinide content and a higher burn-up. Since such fuel would accumulate more He due to alpha damage and alpha decay, consideration must be given to effects on fuel integrity. In addition, the fabrication and reprocessing of such fuel would require higher safety, including criticality control and shielding against neutrons and other radiation sources.

Transmutation systems can be mainly divided into two types: those using power reactors and those using burner reactors specially designed for transmutation. The fuel design concept widely differs between them. The fuel design for the first type is restricted by efficiency and economy as a commercial reactor and therefore fuel having uniform composition yet low minor actinide content would be used. Given the primary requirement of maintaining reactor output at a specified level, it would be difficult to set a high transmutation rate for minor actinides. By contrast, fuel for the second type is designed to fit the reactor for the purpose of maximizing the transmutation rate of the target nuclide. For this reason, fuel would generally be able to have higher content of minor actinides. Such fuel would be placed in a limited location in the core of the burner reactor. However, requirements for fuel are stricter because of a greater change in fuel composition expected during burning.

2.8.4 P&T System

The incorporation of P&T technology into a nuclear fuel cycle would require not only neutron sources, such as the reactor and the accelerator, but also processes for partitioning, fuel fabrication and spent fuel reprocessing. Since transmutation efficiency would not be high enough to complete conversion in a single burning process, a repetition of spent fuel reprocessing and transmutation—or the multiple recycling of minor actinides—would be necessary. This suggests that it would take a significantly long time, like 100 years, to reduce the total amount of minor actinides produced by power generation by means of transmutation. This means that P&T technology must be addressed in a long-term development program for nuclear power generation, such as a reactor design strategy. International studies have been conducted on introduction scenarios for P&T technology [21]. Two major scenarios are shown in Fig. 2.6 [16], which have been studied in Japan: the advanced nuclear fuel cycle using power reactors and the tiered nuclear fuel cycle using burner reactors dedicated to transmutation, as already described.

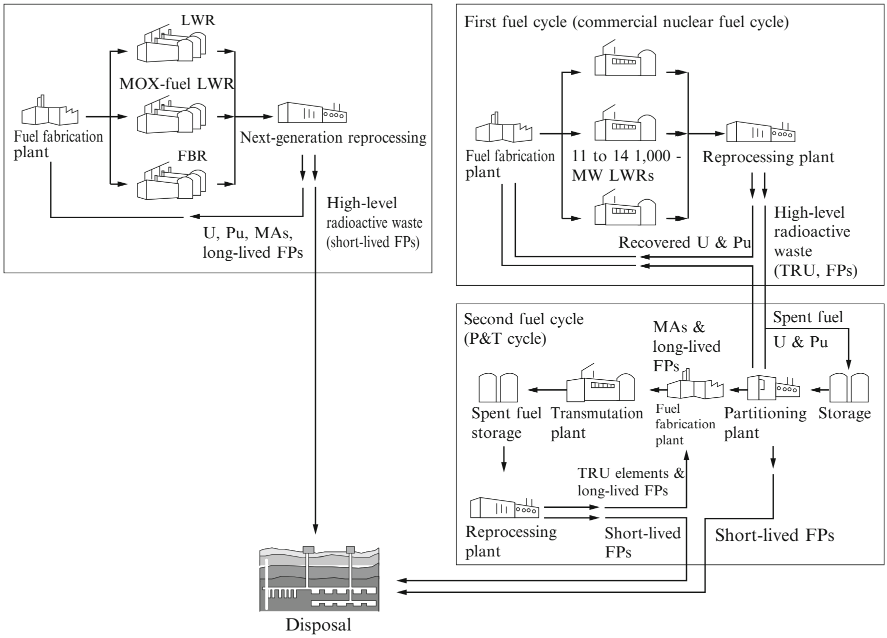

Fig. 2.6 Major partitioning and transmutation systems (*left*: advanced nuclear fuel cycle, *right*: tiered nuclear fuel cycle) [19] – LWR light water reactor, MA minor actinide, MOX mixed oxide fuel, TRU transuranium, FP fission product

In the first scenario, the light water reactor or the fast reactor for commercial power generation is used for minor actinide transmutation, followed by the next-generation reprocessing with an advanced PUREX method for the recovery of minor actinides along with uranium and plutonium. A commercial nuclear fuel cycle serves as a P&T cycle as is, which forms a large-scale transmutation system whereby minor actinides are handled within the nuclear fuel cycle. This is a concept that intends to implement a fuel cycle in a relatively short period of time by advancing existing technologies. As opposed to this, the second concept, the tiered nuclear fuel cycle, seeks to achieve high efficiency transmutation based on newly conceived technologies. In this scenario, minor actinides and other materials used for transmutation are extracted during the partitioning process that follows the reprocessing process in the commercial nuclear fuel cycle, and transmutation is induced in a special plant. Because it combines a separately formed P&T cycle with a commercial nuclear fuel cycle, its whole structure is called the tiered cycle. A benefit of this cycle is the ease of ensuring nuclear safety, such as preventing nuclear proliferation due to the confinement of minor actinides within a small P&T cycle, preventing minor actinides from spreading throughout the commercial nuclear fuel cycle. Most of today's research focuses on the ADS as a special transmutation plant in this cycle.

2.8.5 Wastes from P&T

What kind of wastes would replace high-level radioactive wastes if the P&T system were introduced to the nuclear fuel cycle? Although the amount of long-lived minor actinides contained in high-level radioactive liquid waste from the commercial nuclear fuel cycle would decrease as a result of the introduction of the P&T system, unconventional wastes would be generated from partitioning, transmutation and reprocessing. The adoption of P&T technology would change the flow of materials in the nuclear fuel cycle as well as the kind of wastes generated.

The partitioning and reprocessing processes would generate high-level radioactive wastes that would contain most of the fission products, and secondary wastes including small amounts of actinides and fission products. The loss of actinides would be inevitable in the fuel fabrication process, and activation products and nuclear spallation products would be generated in the transmutation process as radioactive wastes. The wastes expected from the tiered nuclear fuel cycle (Fig. 2.6) are analyzed below for each process.

The 4-group partitioning [22] is assumed as the partitioning process. This separation process divides the elements in high-level radioactive liquid waste into four groups: the transuranic group to be used as fuel for transmutation; the Sr and Cs group including the heat-generating nuclides of ^{90}Sr and ^{137}Cs; the Tc and Pt group for effective utilization; and the group of other elements including iron and rare earth elements. Among these four, the Sr and Cs group and the group of other elements are considered as wastes. Sr and Cs would be adsorbed by titanium oxide and zeolite, respectively, for separation from high-level radioactive liquid waste. These adsorbents, along with adsorbed Sr and Cs, would be burned into a solid, which would then be cemented into metal drums or other containers for conversion into waste form. Vitrification is assumed for the group of other elements. Secondary wastes from this process would be solvent waste and sodium waste; the latter consists of sodium used for solvent washing. Solvent waste would be converted into calcium phosphate by pyrolysis; sodium waste would be converted into sodium nitrate by drying, before finally being solidified.

Transmutation would take place in a special ADS. The accelerator-driven reactor is assumed to have a thermal output of 800 MWt, and lead-bismuth (Pb-Bi) molten salt would serve as both the nuclear spallation target and coolant. The transmutation cycle would be operable only by the electrical output from the ADS. Since fuel would be minor actinide nitride, zirconium nitride would be used as a diluent. The initial load of minor actinides would be 2.5 tons. With the annual transmutation rate of 10 wt%, the system would be capable of transmuting 0.25 tons of minor actinides annually. This amount of minor actinides is equivalent to the amount generated from 195.7 tons of light water reactor spent fuel at 45,000 MWd/t after 4 years of cooling. Considering that JNFL's reprocessing plant can reprocess 800 tons of spent fuel annually, the operation of four ADSs is assumed here. The total reprocessing amount is assumed to be 32,000 tHM, which corresponds to 40 years of operation.

During this transmutation process, nuclear spallation products and activation products would be generated in the nuclear spallation target, and activation products in the coolant. Since the target would double as coolant, these products would be completely mixed and then extracted as dismantling waste when the service life of the transmutation reactor would expire. The amount is estimated to be about 6,500 tons.

The fuel withdrawn from the transmutation reactor would undergo fuel reprocessing, during which the residual minor actinides would be separated and recovered from fission products. It is assumed that this reprocessing would employ the fused-salt electrolysis process, which uses lithium chloride-potassium chloride (LiCl-KCl) salt and liquid cadmium. Major wastes generated here would be salt waste containing the majority of fission products and trace amounts of unrecovered minor actinides; metal waste containing rare fission products, such as platinum group elements; hulls containing activation products; and exhaust including ^{14}C generated from ^{14}N.

Since the fuel cladding, which would be turned into waste hulls, is made of stainless steel, it would be solidified with cement. Rare metal fission products (e.g., Ru, Rh, Pd and Tc) that would not be transformed into chlorides during spent fuel melting would become metal waste and be solidified as a zirconium alloy containing up to 4 % of them. As a means to solidify LiCl-KCl salt waste containing soluble fission products other than rare elements and noble gases, the sodalite solidification method would be used [23].

Figure 2.7 shows the results from analysis of types and generated amounts of radioactive wastes from the above P&T system, which may be called P&T system

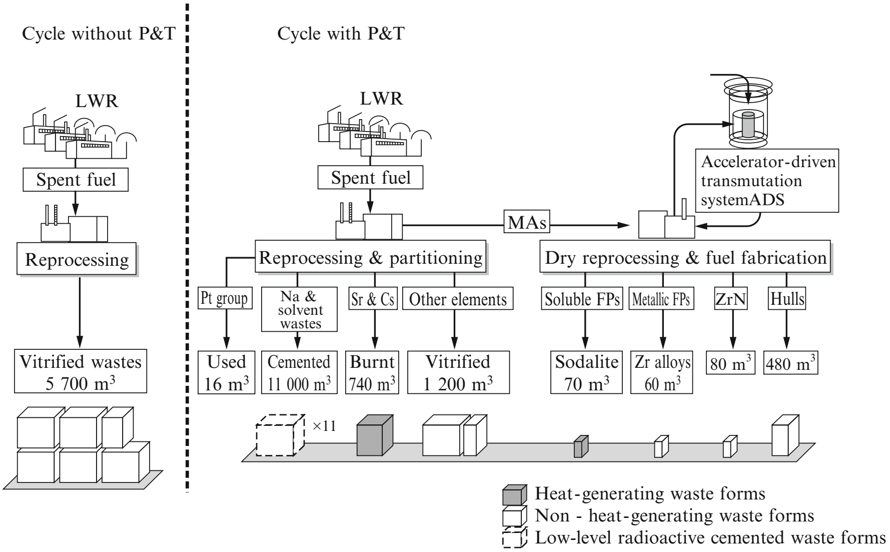

Fig. 2.7 Radioactive wastes from the tiered nuclear fuel cycle [24] –LWR light water reactor, MA minor actinide, FP fission product, ZrN zirconium nitride

wastes and would be generated instead of high-level radioactive vitrified wastes. The analysis is based on the calculation of material balance and radioactivity balance in the P&T system. Among the wastes identified, those with radioactivity levels equivalent to the radioactivity levels of today's high level radioactive vitrified wastes are cemented waste forms produced by burning Sr and Cs generated during the partitioning process; and sodalite waste produced from soluble fission products generated during the reprocessing of transmutation spent fuel. It is suggested that their total volume would be reduced to about 1/7 of that of vitrified wastes generated from the current cycle without P&T. Other wastes would be considered as TRU or low-level radioactive wastes. The type of waste that would particularly be large in generated amount would be the cemented form of the secondary waste (sodium waste and solvent waste) from the partitioning process, which would have radioactivity levels high enough to require concrete vault disposal or subsurface disposal today.

Wastes not included in the chart are operational wastes (e.g., devices replaced during facility operation or maintenance) and facility dismantling wastes. Items contaminated with minor actinides lost during fuel fabrication are not included either. Although activated Pb-Bi coolant (6,500 tons) in the transmutation reactor would be one of the dismantling wastes as mentioned above, studies have not reached the point where quantitative estimations concerning the dismantling of P&T and other related facilities are possible.

All radioactive wastes must be disposed of safely and rationally. P&T technology has the potential to allow high-level radioactive liquid wastes, which would otherwise be vitrified entirely, to be separated according to nuclide properties and thereby enables the design of more rational disposal sites than today's site designs. However, some wastes, like activated Pb-Bi coolant, could raise the issue of chemical toxicity of metals, in addition to being radioactive.

The radioactive wastes addressed in studies on P&T technology is essentially limited to current high-level radioactive wastes. The current nuclear power generation system produces several types of radioactive wastes other than high-level radioactive wastes. Efforts have been made to safely dispose of them by the method suitable for the characteristics of each waste type, such as the types and contents of radionuclides included (Chap. 6). A challenge in the future development of P&T technology is to establish holistic measures for radioactive waste management for the wastes expected from the nuclear fuel cycle after the introduction of P&T.

2.8.6 P&T as a Waste Management Policy

Many countries and international organizations have been eagerly conducting R&D on P&T technology. Japan was one of the first countries to start it, with basic research on partitioning and fast reactor transmutation launched around 1973. In 2009, the Japan Atomic Energy Commission decided that R&D in this area should be continued steadily [25].

With many countries aggressively promoting R&D in this sector, France is the only country that has adopted P&T as a national radioactive waste management policy, putting it on the table for national discussion. To discuss future radioactive waste management policies, France enacted in 1991 the Law of 30 December 1991 Concerning Research Activities on the Management of Radioactive Waste (1991 Law) [26] to stipulate that research should be conducted in three fields: P&T of long-lived radionuclides, and conditioning of radioactive wastes, long-term storage on the ground surface of spent fuel and radioactive wastes, and geological disposal.

In 2005, 15 years later, the results of the research were presented to the French government, which then established the Planning Act on the Sustainable Management of Radioactive Materials and Waste based on them in June 2006 [27]. The greatest advancement from the 1991 Law was that the new Planning Act stipulates that R&D on long-lived, medium- to high-level radioactive waste should principally focus on geological disposal and that P&T and long-term storage should be studied as complementary measures. It is also noteworthy that the French government formulated a clear policy for such R&D with an eye toward commercialization and industrialization. More specifically, with respect to geological disposal, the goal set in the policy is applying for a license to establish a disposal site by 2015 and starting operation by 2025. Regarding P&T, the policy seeks to promote surveys and research on the fourth-generation reactor and the ADS in coordination to complete evaluation of the outlook for industrialization by 2012 and start of operation of a prototype reactor by 2020. Additionally, the policy requires the promotion of long-term storage with the goal of either constructing new storage facilities or modifying existing facilities by 2015 at the latest so that storage capacity and period needs can be met.

In this way, France has been discussing P&T with attention to its potential to contribute to radioactive waste management. The results suggest that the policy on future P&T cannot be considered without addressing the issue of industrialization. Arguments in previous sections show that the introduction of P&T technology has an impact on the technology base for the entire nuclear fuel cycle. The issue is what kind of role this technology should play in the development of the nuclear power generation system over the next century.

Exercises

1. The uranium enrichment process uses, as raw material, natural uranium or uranium recovered through reprocessing, and produces two products: enriched uranium with a ^{235}U content higher than that in natural uranium (0.72 %); and depleted uranium with a ^{235}U content lower than that in natural uranium, which is considered as waste. Answer the following questions about the material balance in the enrichment process.

   (a) Assume that natural uranium is the only raw material input into the enrichment process and that the ^{235}U content in waste is 0.3 %. How many tons of natural uranium are needed to produce 27 tons of 4 % enriched uranium? In this case, how many tons of depleted uranium will be generated?

   (b) Assume that, in addition to natural uranium, uranium recovered through reprocessing is used as a raw material input into the enrichment process. Provided that the ^{235}U content in the recovered uranium is 0.8 % and that 25 tons of this uranium is supplied, how many tons of natural uranium are needed to produce 27 tons of 4 % enriched uranium? In this case, how many tons of depleted uranium will be generated?
   (c) Compare the above two cases and explain the natural uranium saving effect of recycling uranium recovered through reprocessing and its impact on the amount of wastes generated during mining and melting processes.

2. Since the transmutation efficiency of minor actinide transmutation reactors, such as the accelerator-driven reactor, is not high enough to complete transmutation in a single burning cycle, a repetition of spent fuel reprocessing and transmutation—or the multiple recycling of minor actinides—is essential. Answer the following questions, requiring calculation of the amount of minor actinides transferring to wastes during multiple recycling.

   (a) Assume the transmutation efficiency in one burning cycle is α. This means that α, in tons, of minor actinides is transmuted from 1 ton of minor actinides loaded in the transmutation reactor. Then assume β is the ratio of transferring of spent minor actinides discharged from the transmutation reactor to waste through dry reprocessing. Calculate the total amount of minor actinides transferring to waste when 1 ton of minor actinides is transmuted.
   (b) The value of α in a typical accelerator-driven reactor is 10 %. Calculate the migration rate β needed to limit the amount of minor actinides transferring to waste to 1 % or less of the amount before transmutation.

References

1. Radioactive Waste Management Center, *Current State of Research in Uranium Waste Treatment and Disposal*, RWMC Topics, 26 (1993) (in Japanese)
2. Japan Nuclear Fuel Ltd., Japan Atomic Energy Agency, Global Nuclear Fuel – Japan Co., Ltd., Mitsubishi Nuclear Fuel Co., Ltd., Nuclear Fuel Industries, Ltd., and JCO Co., Ltd., *Discussion Paper on Uranium Waste Disposal and Clearance* (2006) (in Japanese)
3. Japan Atomic Energy Commission's Advisory Committee on Nuclear Fuel-Cycle Backend Policy, *Basic Policy on Uranium Waste Treatment and Disposal* (2000) (in Japanese)
4. Nuclear Safety Commission of Japan, *The Clearance Level for Uranium-handling Facilities* (2009) (in Japanese)
5. Radioactive Waste Management Center, *Changes in the Generation and Treatment of Waste from Nuclear Power Reactor Operation*, RWMC Topics, No. 22 (1992) (in Japanese)
6. Japan Nuclear Energy Safety Organization, *FY 2009 Annual Report on Nuclear Facility Operation and Management* (2001), p. 707 (in Japanese)
7. T. Izumi, M. Hagiwara, T. Ohtsu, M. Tsujita, M. Arai, H. Inagawa and M. Nomi, Development of Radioactive Spent Ion-exchange Resin Treatment System, FAPIG, **174**, 40–42, First Atomic Power Industry Group (2007) (in Japanese)

8. Thermal and Nuclear Power Engineering Society (ed.), Water and Chemical Management at Thermal and Nuclear Power Plants – VII. Management of Effluent, Waste Oil and Waste (Nuclear). Therm. Nucl. Power **58**, 49–78 (2007) (in Japanese)
9. Nuclear Safety Division, Safe Environment Department, Fukui Prefectural Government, Operation and Construction Status of Power Plants for March-May 2010, Fukui Prefectural Panel on Nuclear Safety Management (2010) (in Japanese)
10. Radioactive Waste Management Center, *Current State of Treatment, Storage and Disposal Technologies for Reprocessing Waste*, RWMC Topics, 25 (1993) (in Japanese)
11. Radioactive Waste Management Center, *Current State of TRU Waste Treatment*, RWMC Topics, 34 (1995) (in Japanese)
12. Japan Nuclear Cycle Development Institute, FY 2002 Research and Development Subject Evaluation (Interim Evaluation) Report on the Evaluation Subject "Development of Krypton Recovery and Immobilization for Reprocessing Facilities," JNC TN1440 2003–2005 (2003) (in Japanese)
13. T. Sakurai, A. Takahashi, *Behavior of Iodine in the Reprocessing of Spent Nuclear Fuels (Research and Development)*, JAERI-Review 97–002 (1997) (in Japanese)
14. Radioactive Waste Management and Nuclear Facility Decommissioning Technology Center, *Survey on Technologies for the Recycling of Resources Recovered from Radioactive Waste and Reduction of Associated Environmental Burdens Mainly through Nuclide Partitioning and Transmutation* (2006) (in Japanese)
15. Japan Nuclear Cycle Development Institute, *Technological Reliability of Geologic Disposal of High Level Radioactive Waste in Japan – Secondary Report on Geologic Disposal Research and Development (Summary Report)*, JNC TN1400 99–020 (1999) (in Japanese)
16. I. Fujii, *Natural Reactor*, (University of Tokyo Press, 1985) (in Japanese)
17. Japan Atomic Energy Agency, *JAEA Activity Report*, Material for the Fifth Meeting of the Working Group on Research Facility Waste, Committee on R&D in the Nuclear Energy Field, Ministry of Education, Culture, Sports, Science and Technology (May 12, 2009) (2009) (in Japanese)
18. Act on Final Disposal of Designated Radioactive Waste, June 7, 2000, Act No. 117 (2000) (in Japanese)
19. T. Mukoyama, Annihilation disposal of long-lived radioactive waste. Energy Rev. **14**(2), 8 (1994) (in Japanese)
20. Y. Kondo, M. Kubota, T. Abe and K. Nagato, *Development of a Group Partitioning Method: Method of Recovering and Using Useful Elements Contained in Spent Fuel* (literature search), JAERI-M91-147, Japan Atomic Energy Research Institute (1991) (in Japanese)
21. Nuclear Energy Agency of the Organisation for Economic Co-operation and Development, Expert Group, *A Status Report Accelerator-driven Systems (ADS) and Fast Reactors (FR) in Advanced Nuclear Fuel Cycles – A Comparative Study* (2002)
22. Y. Morita, M. Kubota, Wet partitioning and waste treatment. Radioact. Waste Res. **2**, 75 (1996) (in Japanese)
23. T. Nishimura, Y. Itamura, Toward realization of dry recycling technology and a metallic fuel FBR. Denchuken Rev. **37**, 61 (2000) (in Japanese)
24. H. Oigawa, K. Nishihara, K. Minato, T. Kimura, Y. Arai, Y. Morita, S. Nakayama and J. Katakura, *Current State and Future Policy on JAERI's R&D of Partitioning and Transmutation Technology for Long-Lived Nuclides*, JAERI-Review 2005–043, Japan Atomic Energy Research Institute (2005) (in Japanese)
25. Japan Atomic Energy Commission, *Current State and Future Policy on R&D of Partitioning and Transmutation Technology* (2009) (in Japanese)
26. LOI no 91–1381 du 30 décembre 1991 relative aux recherches sur la gestion des déchets radioactifs (91-1381/1991.12.30)
27. LOI no 2006–739 du 28 juin 2006 de programme relative á la gestion durable des matières et d_chets radioactifs (2006-739/2006.6.28)

Chapter 3
Decommissioning of Nuclear Facilities

Taro Shimada

3.1 What Is Decommissioning?

Decommissioning is a series of measures taken after the main activities associated with a licensed nuclear activity have been terminated and before the regulations set forth in the Act on the Regulation of Nuclear Source Material, Nuclear Fuel Material and Reactors [1] (hereinafter referred to as "Reactor Regulation Act") are fulfilled. Decommissioning includes the transfer of nuclear fuel material, removal of contamination caused by nuclear fuel material, and disposal of nuclear fuel material or other materials contaminated with nuclear fuel material. Therefore, the dismantling of facilities, which is undertaken after the main activities have been terminated, is also included in decommissioning. Decommissioning is thus a process to reduce the residual radioactivity of such facilities to the levels necessary for fulfilling the regulations set forth in the Reactor Regulation Act. Because these measures produce various types of radioactive wastes in large amounts in a short period of time, the concept of radioactive waste management needs to be actively incorporated into the planning and implementation of decommissioning. If a decommissioning plan is not adequately formulated, there is a possibility that material that does not need to be handled as radioactive wastes may be improperly classified and handled as such. Furthermore, depending on the dismantling method selected, the amount of secondary wastes generated

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The nuclear regulatory system in Japan has been changed significantly after the Fukushima Daiichi Nuclear Power Station accident in March 2011. Descriptions in this chapter have been translated from the book originally published in Japanese before the accident, with minimal update where appropriate. The Nuclear Regulation Authority newly established after the accident has not completed its review for the guidelines and regulations established by the former Nuclear Safety Commission. In this chapter, guidelines set by the NSC have been adopted.

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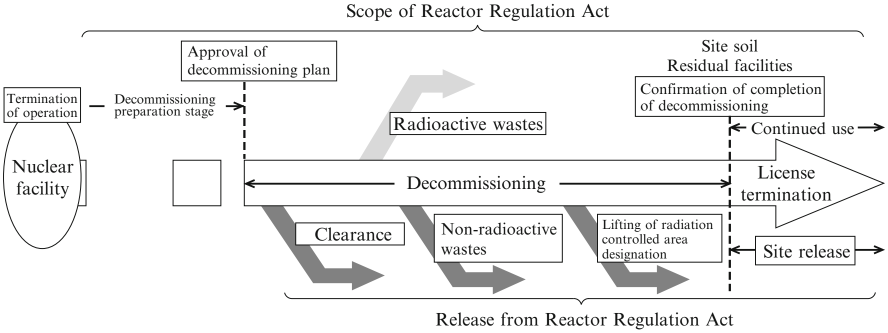


Fig. 3.1 Nuclear facility decommissioning process flow

may increase or decrease and the disposal method for the wastes may also vary. It is therefore important to develop a decommissioning plan based on analytical evaluation, operating history surveys, measurement evaluation and other advance surveys as well as the latest dismantling technology studies. As explained above, there is a close relationship between decommissioning and radioactive waste management.

Instead of leaving radiologically-contaminated facilities or sites without any treatment and possibly affecting the health of the general public in their vicinity, removing residual radioactivity at the earliest stage possible is desirable also from the perspective of reducing the risk of radiation exposure to the public in the surrounding areas. Because removing the radioactivity allows buildings and sites to be used for other activities and thereby benefits society, safe and reasonable decommissioning activities are crucial.

As described in Fig. 3.1, decommissioning involves dismantling nuclear facilities that are regulated by the Reactor Regulation Act, as well as managing radioactive equipment and structures generated in the course of such facility dismantling as radioactive wastes in accordance with the provisions of the Reactor Regulation Act. Items with no history of contamination as well as those decontaminated by physically removing contaminated parts are handled as industrial wastes under the classification of “non-radioactive wastes”. Contaminated items with radioactivity concentrations that meet or fall below the clearance level, which is set as a reference for a sufficiently low concentration level, are released from the regulations of the Reactor Regulation Act by applying the clearance system (see Chap. 4, Clearance) designed to facilitate the effective use of resources and the reduction of radioactive wastes. After equipment is removed from nuclear facilities, the remaining building structures are reused or dismantled as appropriate by applying the clearance, site release or other appropriate systems along with the lifting of the radiation controlled area designation according to operational safety program. The site will also be released from the regulations of the Reactor Regulation Act when the criteria for site release, which will be developed in the future, are met and after the confirmation of the completion of decommissioning measures is obtained from the regulatory authority.

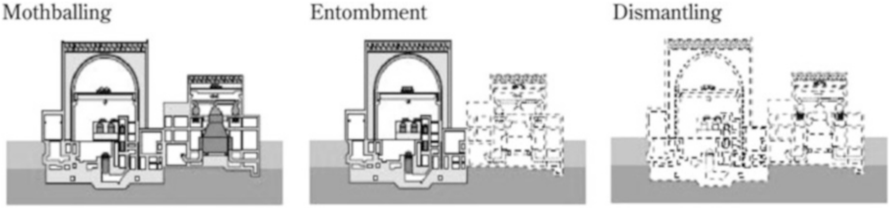


Fig. 3.2 Reactor facility decommissioning methods

3.1.1 Decommissioning Method

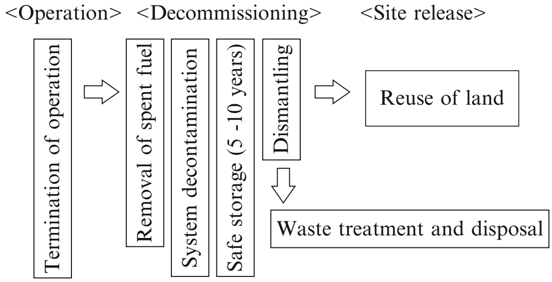
While the methods used for decommissioning reactor facilities vary depending on the siting conditions of the facilities, regulations and standards used in each country, policies adopted by the licensee and other factors, there are three main types of decommissioning methods in general as shown in Fig. 3.2: mothballing, entombment and dismantling.

1. Mothballing: This is a method in which the entire facility is shut down and placed under appropriate control after spent fuel removal and system decontamination are completed. This method is also called safe storage.
2. Entombment: This is a method in which highly radioactive parts of the inside of the reactor building are entombed (openings are sealed by filling with concrete or other materials) and placed under appropriate control after spent fuel removal and system decontamination are completed, and the rest of the facility is dismantled.
3. Dismantling: This is a method in which the entire facility is dismantled after spent fuel removal and system decontamination are completed.

International Atomic Energy Agency (IAEA) classifies immediate dismantling, deferred dismantling and entombment. The U.S. Nuclear Regulatory Commission (NRC) also classifies deferred dismantling (SAFSTOR), entombment as in-situ disposal (ENTOMB), and immediate dismantling (DECON) [2]. In-situ disposal is a method in which a facility is mothballed over a long period of time and subsequently disposed of by burying it on site. This method, however, is no longer a possible option for decommissioning since it was recently decided in the U.S. that decommissioning must be completed within 60 years of the termination of operation. It should also be noted that the concept of "in-situ disposal (ENTOMB)" referred to above differs from that used in the field of radioactive waste disposal.

While all these classifications exist, decommissioning projects in practice are in most cases a combination of mothballing, which is carried out for a certain length of time to allow the radioactive decay, and dismantling, which takes place after the mothballing process.

Fig. 3.3 Standard process of decommissioning



The U.K. and some other European countries recently changed their policy on decommissioning and now intend to reduce risks by dismantling as early as possible facilities that have been mothballed for deferred dismantling. This change was made based on the understanding that while mothballing allows the decrease of residual radioactivity due to radioactive decay, it does little to reduce risks because radioactive materials remain on site during the process.

Meanwhile, Japan's standard approach to the decommissioning of a reactor facility is to dismantle it as soon as possible in principle following the termination of the reactor operation, while maintaining cooperation with the local community and continuing to effectively utilize the land as a site for nuclear power generation, based on the premise of ensuring safety.

The Advisory Committee for Energy of the Agency for Natural Resources and Energy of Japan proposed in 1985 that the standard process for the decommissioning of 1,100-MWe-scale nuclear power plants should involve 5–10 years of safe storage and then subsequent dismantling of facilities over a span of 3–4 years (Fig. 3.3). Based on the proposed standard process, the decommissioning cost was estimated to be about 30 billion yen per reactor. The Reserve Fund for Dismantling Nuclear Power Facilities was set up in 1989, and Japanese electric utilities have since been building up the reserve. The cost of the treatment and disposal of radioactive wastes produced by dismantling commercial nuclear plants was also added to the reserve in 2000. Since its amendment in 2005, the Reactor Regulation Act provides that decommissioning may be started once spent fuel has been removed from the reactor core.

3.1.2 Amount of Wastes Generated from Decommissioning

Large amounts of wastes are generated when a nuclear facility is dismantled. Table 3.1 shows the estimated amounts generated at the nuclear power plants currently undergoing decommissioning. The data indicate that most of the wastes generated from decommissioning nuclear plants are cleared items or non-radioactive waste, which are not required to be handled as radioactive

Table 3.1 Amounts of wastes generated from decommissioning nuclear power plants (unit: ton)

Radioactivity level classification		Tokai Power Station (GCR)	Advanced thermal reactor Fugen	Hamaoka Nuclear Power Station (BWR) Unit 1	Hamaoka Nuclear Power Station (BWR) Unit 2
Low-level radioactive wastes	Waste with relatively high radioactivity concentration	1,530	260	100	100
	Waste with relatively low radioactivity concentration	8,870	1,380	1,000	1,200
	Waste with very low radioactivity concentration	13,080	45,460	6,300	7,900
Wastes not required to be handled as radioactive materials (cleared items)		40,160	510	11,200	13,400
Non-radioactive wastes		128,700	141,000	192,700	249,500

Source: Websites of the Japan Atomic Power Company, Japan Atomic Energy Agency, and Chubu Electric Power Co., Inc.

materials; therefore, radioactive wastes account for only a fraction of the total amount of wastes.

Upon disposing of radioactive wastes, the landfill disposal method is used for wastes with a very low concentration of radioactive materials, while the concrete vault disposal method is used for wastes with a relatively low concentration of radioactive materials. For wastes that are removed from inside the reactors and contain a relatively high concentration of radioactive materials, the subsurface disposal method is envisaged (Chap. 6, Radioactive Waste Disposal). The clearance system, which provides that materials that are generated from nuclear facilities and have extremely low levels of radioactivity concentration are not required to be handled as radioactive materials, was introduced in December 2005 and has been applied to some nuclear power plants and research facilities.

3.2 Formulation of Decommissioning Plan

The licensee of a nuclear facility who intends to decommission the facility is required to draw up a decommissioning plan pursuant to the Reactor Regulation Act and apply for the approval of the regulatory authority. It is required that the decommissioning plan include: the dismantling methods; removal of contamination caused by nuclear fuel materials; disposal of nuclear fuel material or other materials contaminated with the nuclear fuel materials; and the decommissioning process. In addition to the above information to be included in the main text of the document, the decommissioning plan must also be accompanied by supplementary documents explaining the management of radiation exposure; consequences of credible accidents; distribution of contamination caused by nuclear fuel materials and its evaluation method; and facilities and performance that need to retain their functions during decommissioning and the duration required for those functions.

The criteria for the approval of a decommissioning plan are: (1) spent fuel has been removed from the reactor; (2) plans for the management and transfer of nuclear fuel materials are appropriate; (3) plans for the management, treatment and disposal of contaminated materials are appropriate; and (4) plans for the prevention of disasters resulting from the reactor or materials contaminated with nuclear fuel materials are appropriate.

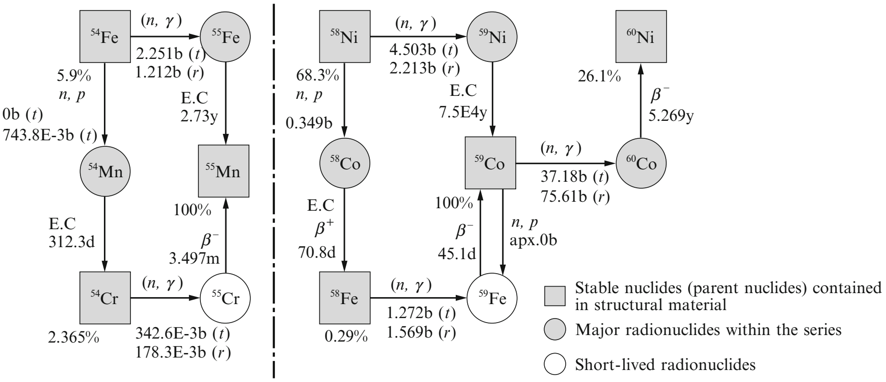
With respect to criteria (3) and (4), the subsequent sections provide detailed explanations on the evaluation of the distribution of contamination caused by radioactive materials, development of a dismantling work plan, management of radiation exposure, and the evaluation of the consequences of credible accidents (safety evaluation). These are considered particularly important among all information to be included in the main text of a decommissioning plan and its accompanying documents.

3.2.1 Evaluation of Residual Radioactive Materials

The evaluation of radioactive materials remaining inside a facility to be decommissioned involves the evaluation of activated radioactivity, which is the source of contamination, and the evaluation of secondary contamination caused by various factors such as the migration of the generated radioactivity or fission products.

3.2.1.1 Evaluation of Activated Radioactivity

Activation is a process in which neutrons generated by nuclear fission or other processes undergo nuclear reactions with coolant, air, or the materials that constitute nearby equipment and structures, and produce radionuclides in them. Radioactivity generated through activation is called activated radioactivity or induced radioactivity. Typical nuclear reactions that cause activation are (n, r), (n, p), (n, $2n$), (n, np), (n, d), and (n, α) reactions. During the operation of a reactor, radionuclides are generated constantly through activation and transform to different nuclides due to radioactive decay. Figure 3.4 illustrates an example of the activation reactions and decay series of a representative element in a metallic structure. To evaluate activated radioactivity, the generation and decay of radionuclides produced through activation are evaluated based on the distribution of the neutron fluence rates, which are obtained by calculation and measurement, inside the operating reactor and its surroundings. The results are then used to determine the concentration and quantitative distribution of radionuclides at a specific point in



Source: Chart of the Nuclides (JAEA) and Radiation Data Book (Chijinshokan Co., Ltd., Tokyo)

Fig. 3.4 Example of reaction series in metallic structure induced by neutron irradiation – b barn, (*t*) thermal neutron capture, (*r*) resonance integral

time. An example of a flow used for calculating activated radioactivity is presented in Fig. 3.5. The three main procedures are described next.

1. Evaluation of neutron fluence rate distribution

The calculation of neutron fluence rates is made in principle by numerically solving the Boltzmann transport equation using the Sn method (discrete ordinate method). The radius of activation induced by neutron irradiation during the reactor operation is limited in part to equipment and structures surrounding the reactor. The general calculation procedure when using transport calculation codes of the Sn method, such as the two-dimensional DORT or three-dimensional TORT, is as follows. First, create the geometric model of the structure subject to evaluation (from the reactor to the biological shield) while creating a neutron macroscopic effective cross section library according to the material composition of each region using nuclear data libraries such as JENDL-4.0 [3] and ENDF/B-VII [4]. Secondly, input source strength data including the neutron energy spectrum in the reactor core as well as the number and distribution of neutrons generated in the entire reactor core area, and set the Legendre expansion order for scattering as well as other calculation parameters for the Sn method. The neutron fluence rate distribution in each mesh region of the geometrical model should then be calculated using transport calculation codes of the Sn method. For areas where neutron streaming occurs, such as ducts and experimental holes, do calculations separately using the Monte Carlo method and make appropriate corrections where necessary.

2. Evaluation of activated radioactivity

Activated radioactivity is calculated using burnup calculation codes, most typically ORIGEN. The amount of radionuclide *i* generated in a specific area (dXi/dt) is calculated based on neutron fluence rate φ using the following equation.

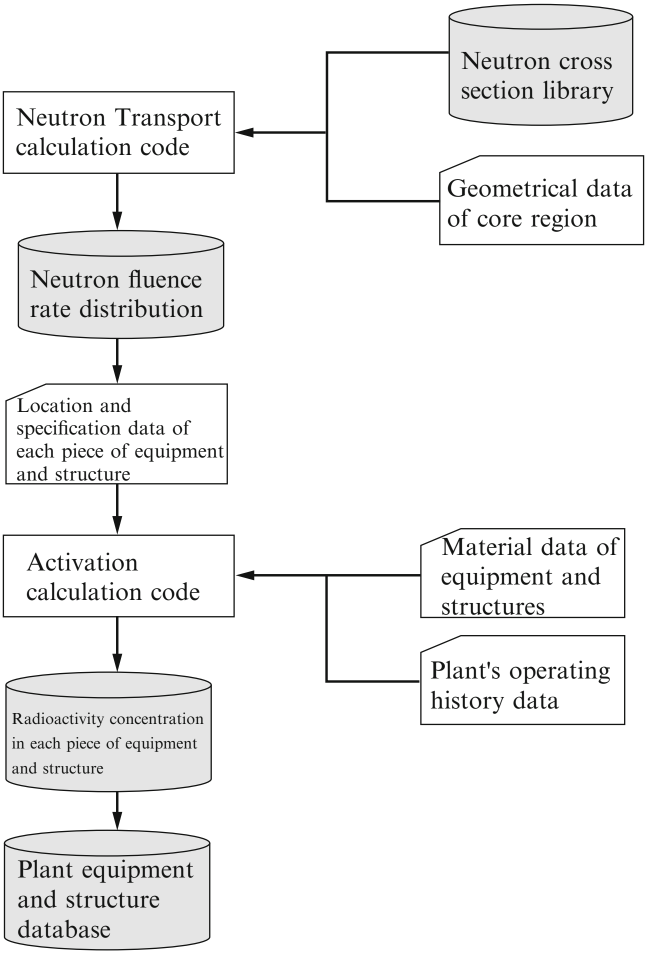


Fig. 3.5 Example of activation calculation flow

$$\frac{dX_i}{dt} = \phi \sum_{k \to i} f_{ik} \sigma_k X_k + \sum_{j \to i} l_{ij} \lambda_j X_j - (\lambda_i + \phi \sigma_i) X_i + y_i F \qquad (3.1)$$

X_i: atom density of radionuclide i generated through activation [n/m^3]
X_k: atom density of target nuclide k [n/m^3]
σ_k (E): neutron absorption cross section of target nuclide k [m^2]
f_{ik}: generation rate of nuclide i when absorbed by nuclide k
λ_i: decay constant of radionuclide i [s^{-1}]
l_{ij}: generation rate of nuclide i when radionuclide j decays

φ: neutron fluence rate averaged in required space integrated over the entire energy range [n/(cm · s)]
σ_i: one-group reaction cross section averaged over spectrum load [m^2]
y_i: fission yield of nuclide i
F: fission rate

The activated radioactivity concentration in each mesh region is calculated by constructing a histogram (hour-thermal output histogram) for the activation calculation based on the operating history of the reactor to be evaluated. For a short-cut calculation, the effective full power year (EFPY) value may be used instead of a histogram. The value is multiplied by the volume of the region to determine the total amount of activated radioactivity contained in the region.

In activation calculations, some trace elements that are negligible when creating the neutron cross section may affect the accuracy of calculations. The amount of impurities contained in a metallic material or concrete may have a significant effect on the calculation results. It is therefore necessary that these values are set properly according to mill certificates or actual measurements.

3. Comparison with measurement results

While the concentration of activated radioactivity can be evaluated by calculation in principle using the above procedure, it is difficult to make an accurate evaluation on the entire system of a large-scale reactor. For such systems, evaluation needs to be made after measuring the radioactivity of samples taken from representative locations, comparing the results with calculated values and making necessary corrections. For example, the results of measurements taken by a neutron detector or gold samples irradiated during operation should be compared with calculation results when evaluating neutron fluence rates. For the concentration of activated radioactivity, comparisons should be made between calculation results and the measurement results of the samples taken from the biological shield by boring machine.

3.2.1.2 Evaluation of Secondary Contamination

Secondary contamination occurs due to various phenomena such as the leakage of nuclear fuel materials or fission products from the fuel, the release of fission products from uranium adhered to the surface during fuel manufacturing, the dissolving of metals in structural materials, the activation of structural materials, or the deposition or peeling of structural material surfaces where in contact with liquid. The contamination spreads over the entire system including equipment and piping. If a leakage of liquid or gas containing radioactive material occurs, the material may adhere and deposit on the surfaces of building structures and cause surface contamination. The level of contamination caused by radioactive material on the inner surface of the system in contact with liquid varies depending on the operating history of the facility, fuel conditions and other factors. For this reason, the evaluation is usually made using the values actually measured on site although attempts are being made to evaluate the level of contamination using calculation codes based on the radioactivity balance. For example, because the compositions of radionuclides found inside pipes carrying coolant with common characteristics, such as those in a reactor cooling system, are mostly the same, the level of radioactivity contamination remaining in each system is evaluated by combining sample collection at representative points and the measurement of dose rates taken outside the pipes.

3.2.2 Development of a Decommissioning Plan

In order to carry out decommissioning smoothly, important steps include formulating a dismantling plan, estimating the number of workers required, and evaluating necessary costs before work is commenced. Upon preparing the dismantling plan, it is recommended that evaluation be made in advance on the characteristics (weight, shape, material, amount of radioactivity, location, etc.) of the equipment and structures present in the facility and a database be created to allow necessary information to be retrieved efficiently. Dismantling work for a nuclear facility consists of a combination of various steps including dismantling using conventional methods, remote dismantling, decontamination and measurement of radioactivity in buildings, and the dismantling of buildings. Each of these steps also involves preparation, cutting, storage and cleanup work. In the planning, these work steps are broken down to formulate a work breakdown structure (WBS). A decommissioning plan is developed by first identifying all necessary work steps as well as determining work conditions and procedures, followed by the restructuring of the work steps. This will allow relevant management data to be calculated including the number of required man-hours, exposure doses of workers, costs, and the amounts of wastes generated, thereby facilitating the development of a rational plan.

One of the tools developed to assist the formulation of a practical decommissioning plan is a Code System for Management of Reactor Decommissioning (COSMARD) [5], a computer program that uses a work package in which work configuration and conditions necessary for the dismantling work are assigned as the input conditions. And it calculates management data for each basic work step (work unit) or each set of such work steps based on information prepared in advance on the physical quantities and residual radioactive materials of the equipment and structures used in the facility (physical inventory database), dose rates of the work areas (work environment database) and calculation models for the management data (work unit database). The work unit database is an aggregation of calculation models written in a simplified language including work unit factors, which indicate the number of man-hours required for dismantling work for each unit weight. Management data associated with the dismantling of equipment and structures are calculated using calculation formulas and numeric values.

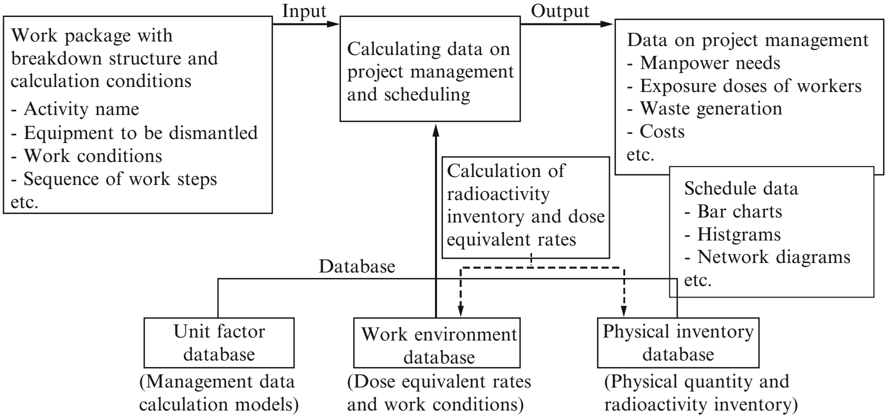


Fig. 3.6 Concept of COSMARD [5]

The concept of the management data calculation using COSMARD is described in Fig. 3.6. COSMARD consists of a management data calculation program as well as an auxiliary program to calculate residual radioactive materials and dose rates, which are necessary for calculating management data. Regarding the work configuration required as part of the input conditions, a work unit or a set of work units can be written using the work breakdown structure. In addition, work conditions can be assigned to each work unit individually where applicable.

3.2.3 Evaluation of Safety

During dismantling work, radioactive gases or particulates may be released into the work environment due to operations such as cutting of objects contaminated with activated radioactivity or radioactivity from secondary contamination. Some of these gases or particulates may be carried through the filters of the exhaust system of the facility building and released into the atmosphere from the ventilation stack. When liquid is used to cut or treat objects, radioactive materials may be transferred to the liquid phase, and some may be discharged into the ocean from the outlet of the facility as liquid waste through the drainage system.

These substances can cause radiation exposure of the general public in the vicinity of the facility. When developing a decommissioning plan, safety must be confirmed in advance by evaluating the expected exposure dose for each exposure pathway through the calculation of the amount of radioactivity released each fiscal year based on the residual radioactivity inventory as well as the types and processes of dismantling work.

Radioactive materials may also be released into the atmosphere in a short period of time if an accident such as a fire or explosion occurs during cutting or other operations that use a flame. Safety must be confirmed in advance by identifying accidents that may occur during decommissioning and calculating the amount of radioactivity released into the atmosphere in the event of such accidents to evaluate the exposure doses of the general public in the vicinity of the facility. It is also necessary to calculate the expected collective exposure doses based on the work plan to ensure that the decommissioning plan will limit the radiation exposure of workers engaged in the dismantling work to the level as low as reasonably achievable.

3.2.3.1 Evaluations of the Exposure Doses of the General Public in the Vicinity of the Facility at Normal Situation

These evaluations can be classified into two steps: released radioactivity evaluation, which covers the period to the release of radioactive gases or airborne particulates into the environment due to operations such as the cutting of equipment or structures; and exposure dose evaluation, which assesses the exposure doses expected after the release of radioactivity.

Released radioactivity evaluation is done for the assumed dismantling work including the cutting and decontamination of specific objects and the storage of the object into waste containers. It also includes the assumed radioactive particulates generated during the dismantling work that are transferred to the gas or liquid phase, and that pass through filters or leak from a contamination control enclosure or building and are released into the atmosphere or ocean. Figure 3.7 illustrates the atmospheric discharge pathways of radionuclides. Released radioactivity is evaluated separately for each of these pathways. Transfer to the gas phase is evaluated taking into consideration the dispersion rate of each nuclide and the collection efficiency of filters. As the inside of the building is usually maintained under negative pressure at normal situation, the leakage from the building is considered negligible and the leakage rate is therefore set as zero in the calculations. In the case of dispersion to the liquid phase, the amount released into the ocean is evaluated taking into consideration the decontamination factor of each nuclide by the liquid treatment system.

The evaluation of exposure doses expected following the release of radioactivity estimates the amount of surface deposition of radionuclides released into the atmosphere using meteorological data such as wind direction and speed for each bearing and taking into consideration radioactivity concentration in the atmosphere and surface deposition velocity. The external and internal exposure doses of the general public in the vicinity of the facility are then calculated for each exposure pathway.

Furthermore, exposure dose from direct and skyshine radiation caused by temporarily stored waste containers with dismantled objects is also evaluated by calculating the amount of the containers generated and taking into consideration the layout of the containers inside the building. The series of evaluations is coordinated with work processes and conducted each fiscal year.

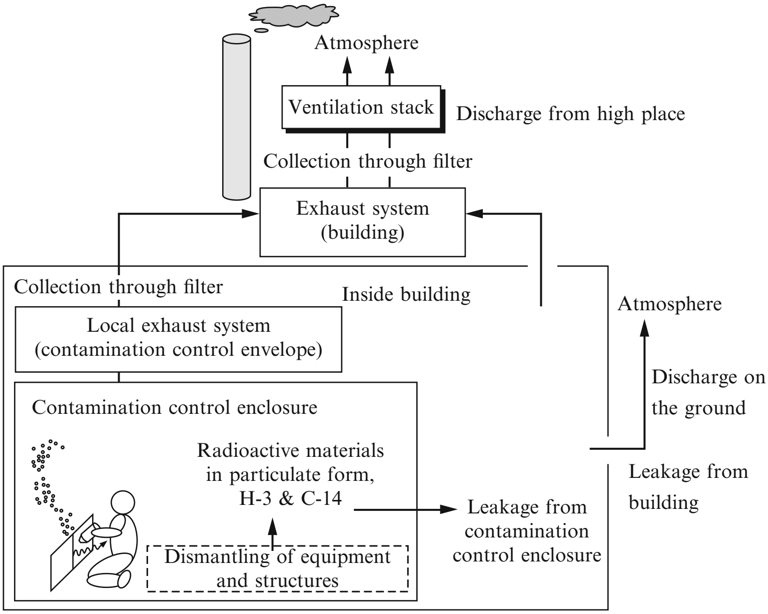


Fig. 3.7 Atmospheric discharge pathways of radionuclides at normal situation

3.2.3.2 Evaluation of the Exposure Doses of the General Public in the Vicinity of the Facility at an Accidental Situation

Table 3.2 lists postulated events at the time of an accident during decommissioning as well as the locations of major mobile inventories. A mobile inventory is the mass of radioactive materials that may possibly be dispersed in the work environment due to combustion or other phenomena when an event occurs. Because combustion temperature is low in a normal fire, the dispersion of radioactive gases or airborne particulates caused by the melting or evaporation of activated base metal is not factored into the evaluation.

This evaluation is performed in the order of: (1) calculation of each mobile inventory accumulated according to the types of dismantling work; (2) calculation of the amount of radioactivity released into the atmosphere based on the mobile inventory expected when a postulated event occurs and the dispersion rate of the mobile inventory into the air; and (3) evaluation of the exposure dose of the general public in the vicinity of the facility for each exposure pathway.

In (1), some of the results of the evaluations conducted on released radioactivity at normal situation are used to calculate the amount of accumulated or deposited mobile inventory that may disperse into the air when a postulated event occurs. The calculation is made separately for each object to be dismantled, taking into consideration the collection efficiency of the filters and the adherence and sedimentation of radioactive particulates. More specifically, evaluation is made by referring to the

Table 3.2 Postulated events and locations of major mobile inventories at the time of occurrence of each event

Classification	Postulated events	Locations of major mobile inventories
Fire	Local filter fire	Inside of filter (immediately before filter replacement)
	Building filter fire	Inside of filter (immediately before filter replacement)
	Contamination control enclosure fire	Adhered to inner walls of a contamination control enclosure or objects to be cut
	Fire from cardboard boxes for temporary storage	Temporary waste storage
Explosion	Unplanned explosion of explosives intended for controlled blasting	Wastes, etc. associated with temporary storage near explosive storage area
	Accidental explosion during controlled blasting operation	Biological shielding concrete
	Explosion of flammable gas	Adhered to inner walls of a contamination control envelope
Filter damage	Local filter damage	Inside of filter (clogging)
	Building filter damage	Inside of filter (clogging)
Loss of electric power	Loss of external power supply	Airborne radioactive material in work space
Drop	Waste container drop	Dross from cutting operation, waste ion exchange resin, etc.
	Cut piece drop	Pieces from cutting activated or contaminated equipment, etc.
	Fuel assembly drop	Spent fuel in transit

general specifications of HEPA filters and setting the maximum amount of mobile inventory that can be held by filters in a usable condition, as well as the amount held by filters when mobile inventory is accumulated to the point that filters are damaged by rising differential pressure. The amount of accumulated radioactive particulates at the point that the above set amount is reached is then calculated separately for each nuclide.

In (2), the total mobile inventory where an accident has progressed to the final stage is calculated. The amount of radioactivity released into the atmosphere in a short period of time during such an accident is also calculated for each nuclide and mobile inventory.

Calculations in (3) are made independently from the evaluation method that uses statistically processed meteorological data for normal situation. This step calculates external exposure to radiation caused by radioactive gases, airborne particulates or other substances in the atmosphere, as well as internal exposure doses from inhalation of such radioactive materials, after radioactivity is released into the environment as a result of a certain event. Out of 8,760 relative concentration and dose values calculated based on meteorological observation data recorded for each hour according to the meteorological guide [6] for the time of an accident, values that represent 97 % annual cumulative frequency of occurrence are applied to these calculations.

3.2.3.3 Exposure Doses of Workers

A collective external exposure dose (D_{ex} [Sv]) of workers engaged in dismantling work is calculated by multiplying the average dose rate (R_{ijk} [Sv/h]) at the position of a worker of each occupation by work hours (T_{ijk} [h]), and is presented as follows.

$$D_{\mathrm{ex}} = \sum_{i=1} \sum_{j=1} \sum_{k=1} R_{ijk} \cdot T_{ijk} \tag{3.2}$$

i: occupation (worker, supervisor, technical supervisor, radiation control manager, etc.)
j: dismantling work classification (preparation, dismantling or cleanup)
k: daily work classification (entry to or exit from area, preparation, dismantling (main or assistance), cleanup, etc.)

The average dose rate at the position of a worker of each occupation changes with the progress of the dismantling work, and such changes vary depending on the layout of equipment and the order of removing the equipment. In normal cases, equipment having a high dose rate is removed first to reduce worker exposure. However, in cases where such equipment is not easily accessible, other pieces of equipment located around it are dismantled first. In many cases the work is performed in crew units consisting of multiple occupations such as a supervisor, workers and a radiation control manager. While workers undertake the cutting operation near equipment, the supervisor is responsible for providing directions and assistance from a location where the entire operation can be observed. A different dose rate therefore needs to be set for each different occupation.

The position of a worker in a work area and the amount of time spent therein vary depending on the worker's occupation and the classification of work performed. Work classifications within a work process are preparation, dismantling and cleanup, while daily work classifications consist of work units such as entry to or exit from the area, preparation, dismantling and cleanup. Work hours for each work unit and effective work hours for each occupation taking into account the allocation of time in a day can be evaluated using the WBS formulated in Sect. 3.2.2, "Development of a decommissioning plan."

Meanwhile, a collective internal exposure dose (D_{in} [Sv]) is calculated by multiplying the radioactivity concentration in the air inhaled by a worker (C_{ijk} (t) [Bq/m^3]) by work hours (T_{ijk} [h]), breathing rate (R_{ik}) and effective internal exposure dose conversion factor [7] for each nuclide (A_m [Sv/Bq]), and is presented as follows.

$$D_{\mathrm{in}} = \sum_{i} \sum_{j} \sum_{k} C_{ijk}(t) \cdot T_{ijk} \cdot R_{ijk} \cdot \frac{1}{W_p} \cdot A_m \tag{3.3}$$

W_p used in the above equation indicates a protection factor, which is a number quantifying the effectiveness of protective equipment. For example, the protection factor is 10 for a half-face respirator, 50 for a full-face respirator and 2,000 for a ventilation suit.

Radioactivity concentration in air at the work space changes significantly in terms of space and time in the course of each operation. The evaluation of radioactivity concentration in air must be conducted applying area and hours that are accurate in light of the actual conditions of each operation. For example, when evaluating the exposure of workers engaged in the cutting operation inside a contamination control enclosure based on the radioactivity concentration in air, calculation should use the average radioactivity concentration of the small area around the cut section. Evaluation for workers not engaged in the cutting operation should use the average radioactivity concentration in air in the entire contamination control enclosure. Similarly, the amount per unit of time of radioactive gases and particulates, which are generated and dispersed in the air due to the cutting of equipment or structures, is dependent on the kerf width and cutting speed of the method used; therefore, data that correspond to the actual cutting conditions must be prepared in advance of evaluation.

3.3 Decommissioning Techniques

3.3.1 Decontamination Techniques

Decontamination conducted upon decommissioning is an effective technique for reducing worker exposure during dismantling work as well as for reducing radioactive wastes. Decontamination can be divided into system decontamination and decontamination after dismantling depending on the timing of the work applied.

Objects subject to decommissioning may be of various materials and shapes, and can have various forms of contamination. There are also many decontamination techniques based on different principles. Optimum techniques should be selected when carrying out decontamination, taking into consideration the purpose of the decontamination work, characteristics of objects to be decontaminated and the cost-effectiveness of using each technique. Representative decontamination techniques are listed in Table 3.3.

An overview of decontamination techniques applicable to decommissioning is given below for system decontamination and decontamination after dismantling depending on the timing of work applied.

3.3.1.1 System Decontamination

While system decontamination is a method also used during service, it is used in decommissioning prior to dismantling work to remove contaminants attached to the inner surface of the system by supplying a decontamination solution into piping that

Table 3.3 Representative decontamination techniques

Object	Classification	Principle	Decontamination techniques
Metallic equipment or structures	Physical	Mechanical method	Brush polishing, grinding, wiping
		Jet method	Steam, high – pressure, ultrahigh – pressure
		Blast method	Dry ice, CFC, water, ice, metallic particle (steel, zirconia, alumina), sponge, etc.
		Vibration method	Ultrasound, vibrating machine
		Laser method	YAG, excimer, CO_2 lasers
		Other	Microwave, melting, strippable coating, thermal shock
	Chemical	Immersion method	Acid (HCl, H_2SO_4, HNO_3, HF, etc.), alkali, oxidant, reductant
		Foam method	Chelator, surfactant, corrosion inhibitor
		Gel method	Combination of Gallant and acid (HCl, H_2SO_4, HNO_3, HF, etc.)
		Paste method	Decontamination agent + filler + carrier
		Other	Formic acid decontamination method (formic acid for reductive dissolution)
			NP – LOMI method (potassium permanganate for redox dissolution, formic acid, vanadium, picolinic acid)
			CORD method (permanganic acid for redox dissolution, oxalic acid, hydrogen peroxide solution UV)
			DfD method (fluoroboric acid for redox dissolution, potassium permanganate)
			Cerium (IV) decontamination method (cerium sulfate for oxidative dissolution)
	Electrochemical	Electrolytic method	Electrolytic polishing, dilute sulfuric acid, phosphoric acid, etc.
	Combined	Electrochemical & physical	ECB method (Electrolytic polishing and mechanical polishing)
		Chemical & physical	Chemical solution and vibration (ultra-sound and machine)
		Chemical & electrolytic regeneration	Redox ($Ce^{3+} \rightarrow Ce^{4+}$), hydroxyl radical ($Ag^{+} \rightarrow Ag^{2+} \rightarrow \cdot OH$)
Concrete surface	Cutting	Mechanical	Milling cutter (planer), grinder
		Cutting	Water jet, blast (several types of abrasives)
	Crushing	Impact crushing	Scabbling, breaker
		Splitting	Spaller
	Thermal		Laser, microwave
	Chemical		Chemical dissolution, electrochemical, biochemical, and coating methods

forms a single loop, such as of a reactor cooling system. This will significantly reduce the dose rate around the piping, which can help reduce the level of worker exposure. The representative techniques include the LOMI method, which uses reductive dissolution, and the CORD method, which uses oxidation-reduction (redox) dissolution. When a facility is in operation, the use of powerful decontamination solutions that may have serious effects on the base metal is avoided in order to maintain the equipment performance. On the other hand, because such effects do not need to be considered during decommissioning, the cerium (IV) decontamination method and the DfD method, which can dissolve base metal, are effective. However, there may be some variations in the effectiveness of system decontamination as a result of the effects of the oxide film on the surface layer or the flow conditions inside the system. Another disadvantage is a large volume of secondary waste generated from filling decontamination solutions in the piping and other parts of the system, necessitating the treatment of decontamination solutions and disposal of secondary wastes.

3.3.1.2 Decontamination after Dismantling

Contrary to system decontamination, decontamination after dismantling is a method carried out after equipment is removed from its original position. Techniques include physical, chemical, electrochemical and thermal decontamination. Decontamination after dismantling does not have a substantial effect in reducing worker exposure; it is rather effective in lowering the classification of radioactive wastes by one or more levels. For example, a radioactivity concentration of waste subject to concrete vault disposal without decontamination may possibly be reduced to the level only requiring landfill disposal after decontamination, thereby reducing the cost of disposal. It is important to fully understand the surface contamination density and characteristics of contamination in advance and determine to what level the surface contamination density can be reduced by decontamination after dismantling.

While the immersion method is used for chemical decontamination in general, this method requires flow of the decontamination liquid. It is also difficult to remove both the oxide film and base metal together, and additional treatment may be needed to dissolve the base metal. The blast decontamination and polishing methods can remove the base metal through physical grinding and are therefore effective for decontamination after dismantling; however, contamination is transferred to the blasting material or the abrasive, for which treatment or disposal needs to be arranged. Laser decontamination is capable of removing the oxide film and base metal at one time by turning up the output level, and is very effective in treating localized contamination. Nevertheless, it is inefficient in decontaminating a wide area in light of its treatment area and rate, which leaves room for improvement in its applicability.

3.3.2 Remote Techniques

In nuclear facility dismantling, the facility may have some areas that cannot be accessed easily due to equipment or structures highly activated or contaminated with radioactivity. Work in these areas are carried out by using remote devices or remote operations such as underwater cutting of equipment, which is performed while equipment is placed in water used as shielding material.

Remote devices employed in dismantling work in the past have included a robot manipulator type, mast (rotating pillar) type, and mast-arm type, which is a combination of the first two. A manipulator with a mast allowing rotation, upward and downward movements and multiple degrees of freedom is attached with a cutting device to its tip to perform dismantling operations. There are several methods of controlling a robot manipulator, such as by sending real-time commands for rotation angle of 6–7 axes from so-called master-slave type equipment, or by specifying the locus of the coordinates of the tip in advance on the computer for adjustment of automatic standoff (distance between the tip of the torch and the surface of the object to be cut) and attitude control to ensure optimum cutting performance. The most desirable method is selected after considering cost-effectiveness and other factors concerning each method.

In the underwater cutting of reactor core internals, dross generated from the operation increases the turbidity of water, leading to reduced visibility and increased radioactivity concentration in the water. Particulate matter floating in the water needs to be collected by filters or other means to prevent turbidity, and measures must be prepared to protect workers from exposure while engaging in this process.

There are also other technologies under development, including a mouse-like robot that has a built-in radiation detector and is able to move freely inside piping by remote control to measure the distribution of contamination inside piping.

3.3.3 Dismantling Techniques

Representative dismantling techniques are listed in Table 3.4. Cutting methods used in dismantling work are classified broadly into thermal cutting methods (plasma arc, laser, oxidation reaction heat methods, etc.), which are intended for metallic equipment and structures, and mechanical cutting methods (abrasive water jet method, nibbler, shearing machine, wire saw, disk cutter, etc.), intended for both metals and concrete. Blasting techniques (shaped charge pipe cutting, controlled blasting of concrete, etc.) as well as electric discharge machining techniques for cutting metals are also used in dismantling operations.

While thermal cutting provides relatively high cutting rates leading to high levels of work efficiency, processes involve the melting and evaporating of base metals, which produce large amounts of airborne particulates in the work environment. In cases where an object to be dismantled has a high radioactivity concentration or surface contamination density, measures to prevent the spread of

Table 3.4 Representative dismantling techniques

Object	Classification	Principle		Dismantling techniques
Metallic equipment or structures	Thermal cutting	Laser		CO, CO$_2$, YAG, and iodine lasers
		Electric	Plasma	Plasma arc
			Arc	Arc saw, TIG, MIG, consumable electrode water jet
		Combination of electric and oxidation reaction heat		Oxygen gas, powder/gas, oxygasoline torch, thermite reaction lance
		Oxidation reaction heat		Oxy-arc, Combination of gouging and gas (G&G method)
		Electric discharge		Electric discharge machining (EDM)
		Metal disintegration		Metal disintegration machining (MDM)
	Mechanical cutting	Grinding		Abrasive water jet, abrasive
		Physical	Reciprocating motion	Nibbler, shearing machine, hacksaw, reciprocating saw, guillotine saw, milling cutter
			Rotational motion	Band saw, chain saw, diamond wire saw, disk cutter, carbide tipped circular saw, roller cutter, pipe cutter, hinge cutter, clamshell lathe
		Blasting		Shaped charge
Concrete structures	Cutting	Mechanical		Diamond wire saw, core boring, disc cutter
		Cutting		Abrasive water jet, shaped charge, liquefied gas
	Crushing	Impact		Breaker, steel ball
		Shock		Controlled blasting
		Pressure		Crusher, jack, lock jack, burster
		Dilatational deformation		Static crushing agent, shape memory alloy
	Heat	Heat		Flame cutting, thermite, laser, electromagnetic induction, and direct electrification methods

contamination as well as protection measures for workers, such as ventilation suits, are necessary.

3.4 Dismantling of Nuclear Facilities

Methods used for dismantling a nuclear facility vary depending on the type and size of the facility. The following sections introduce actual cases of dismantling, with separate discussions for reactor facilities and nuclear fuel cycle facilities.

3.4.1 Dismantling of Reactor Facilities: Examples

A reactor facility, while in operation, continuously generates neutrons in its core through fission reactions, and there are typically large amounts of contamination from activation around the reactor at the time of decommissioning. Of all the radionuclides produced by activation, the radioactivity of short-half-life nuclides decays in a short period of time following the termination of reactor operation. Therefore, by ensuring an adequate cooling period length with safe storage, the exposure dose of workers engaged in dismantling work as well as the amount of radioactive wastes can be reduced.

Two representative examples of the dismantling of reactor facilities are described below.

3.4.1.1 Japan Power Demonstration Reactor [8]

The Japan Power Demonstration Reactor (JPDR) owned by the former Japan Atomic Energy Research Institute's Tokai Research Institute was Japan's first BWR for power generation (initial thermal output 45 MW; post-reconstruction output 90 MW; electric output 12.5 MWe); it reached its initial criticality on August 22, 1963, generated its first electrical power on October 26 of the same year, and ceased operation in March 1976.

The dismantling of the facility started in December 1986. After first removing the dump condenser equipment (equipment used to condensate steam that had doubled in volume after reconstruction) to secure temporary storage space for wastes, the dismantling of equipment inside the reactor containment vessel was commenced. In order to reduce the external exposure of workers and minimize the amount of radioactive gases and particulates released into the environment, remote handling devices and underwater cutting methods were effectively used to dismantle highly radioactive equipment with existing ventilation system.

The dismantling of equipment in the reactor containment vessel was undertaken in the following order. (1) Equipment located around the containment vessel was removed to secure space for installing remote devices such as master-slave type manipulators and mast type devices. (2) After remote handling devices were installed, reactor core internals were dismantled by underwater plasma arc cutting. The reactor pressure vessel was dismantled using the underwater arc saw cutting technique. (3) The dismantling of biological shielding concrete was carried out using techniques such as the core boring method, diamond saw method, water jet cutting device and controlled blasting method. (4) Remaining equipment was removed, and the floors, walls and ceiling planes were decontaminated using scabblers and other devices. (5) Overall measurements were conducted to confirm the completion of decontamination, and the designation of radiation controlled area was lifted. (6) The reactor containment vessel was demolished using conventional techniques.

The amount of radioactive wastes generated during the JPDR dismantling was 3,770 tons. Metals and other radioactive wastes having low radioactivity concentrations were stored in 200-liter drums or 1-m^3 or 3-m^3 steel containers. Highly activated metallic equipment was stored in designated shielding containers specially fabricated for the purpose. On-site shallow landfill disposal was selected for concrete fragments amounting to 2,000 tons, which had a very low radioactivity concentration, as a buried disposal verification test (see Fig. 6.3 in Chap. 6). The number of man-days expended to complete the JPDR dismantling work was 145,000. The collective exposure dose of the workers was 0.3 man-Sv, which was 1/3 of the planned value.

3.4.1.2 Shippingport Atomic Power Station [9]

The Shippingport Atomic Power Station in the U.S. was a PWR built for power generation with an electric output of 100 MW. Its operation began in 1957 and was terminated in 1982. The plant also went through reconstruction during the course of its operation. Decommissioning started in 1985 and was completed in 1989, during which the reactor pressure vessel was removed as an assembly without being cut or separated into segments. This approach was employed after it was decided that removing the reactor pressure vessel assembly as a whole, instead of cutting it in parts and storing them in waste containers, would provide more benefits such as reduced cost, shorter term of work and lower worker exposure. As shown in Fig. 3.8, the pressure vessel was placed in the outer neutron shield tank, and reactor core internals, filters and other elements were stored in the pressure vessel. This assembly was then processed into a waste form, and was shipped long-distance (Fig. 3.9) through land transportation by truck trailer and water transportation by a carrying vessel to the low-level radioactive waste burial site located in Hanford, Washington, where it was buried for disposal. The decommissioning cost was about 90 million dollars, and the population exposure dose of the workers was 1.55 man-Sv.

3.4.2 Dismantling of Nuclear Fuel Cycle Facility: Examples

Unlike in reactor facilities, neutron irradiation of equipment and structures does not need to be considered in nuclear fuel cycle facilities and there is no contamination from activation. Radionuclides used in nuclear fuel cycle facilities have a long life, and it is not necessary to allocate a specific time interval between the termination of facility operation and the start of dismantling work because radioactive decay cannot be expected sufficiently. A nuclear fuel cycle facility consists of many small caliber pipes, small equipment, tanks, vessels and other components, and its systems have complex structures. Large amounts of nitric acid-resistant stainless steel as well as Ti and Zr alloys are often used as component materials. The

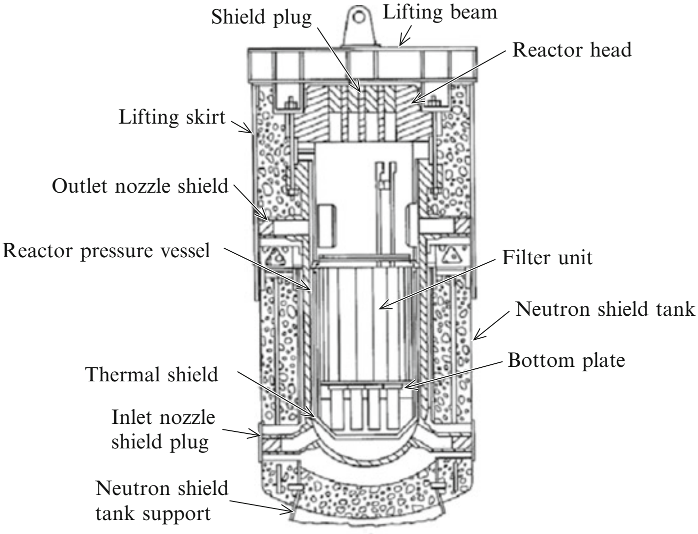


Fig. 3.8 Configuration of the waste form of the Shippingport reactor pressure vessel removed as an assembly [10]

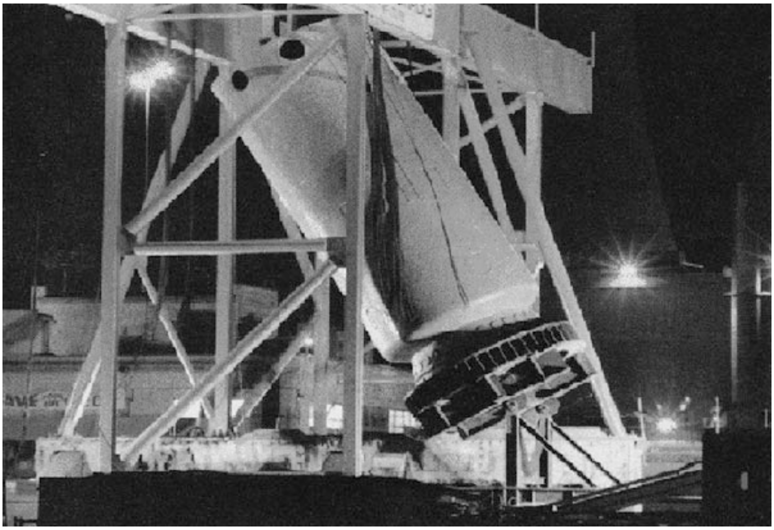


Fig. 3.9 Waste form of the Shippingport reactor pressure vessel being carried out of the facility [11]

dismantling work environment includes many narrow areas, where radionuclides of uranium and transuranium such as plutonium are used in various forms including gases, liquids and powders. The work must be conducted by selecting appropriate methods according to the types of radionuclides.

Three representative examples of the dismantling of nuclear fuel cycle facilities are described below.

3.4.2.1 Dismantling of Hanau Fuel Fabrication Plant [12]

Siemens AG, based in Germany, decommissioned its uranium and MOX fuel fabrication plants and hot cells that had been constructed in Hanau. The Hanau MOX fuel fabrication plant was in operation from 1965 to 1991, where 26,000 MOX fuel rods were fabricated from 8.5 tons of plutonium. After the operation of the plant was terminated, the treatment of residual nuclear fuel materials was carried out until early 2001, and the dismantling of the fabrication equipment followed. Two hundred forty glove boxes installed in the plant were dismantled and disposed of during the process. Radioactive wastes generated from dismantling the plant were solidified through cementation and have been stored in containers that met the burial conditions of the Konrad repository. The dismantling work was completed in September 2006, and the site was released.

3.4.2.2 Reprocessing Test Facility [13]

The Japan Atomic Energy Research Institute (predecessor of the current Japan Atomic Energy Agency) Reprocessing Test Facility (JRTF) was constructed in 1966 as Japan's first engineering-scale reprocessing research facility that studied fuel reprocessing based on the PUREX method. It conducted aqueous reprocessing tests between 1968 and 1969.

Upon dismantling installations and equipment, the surface contamination density and dose rates of the work area were measured first, and wipe decontamination, contamination fixation and radiation shielding were applied where needed. Based on the degrees of the contamination of the installations and equipment, contamination control enclosures with up to four compartments and local exhaust systems were installed to prevent radioactive particulates and other material from leaking outside the work area.

For dismantling glove boxes and large tanks, mechanical cutting tools such as band saws, pipe cutters and nibblers were mainly used as appropriate taking into consideration the material, size, structure and other characteristics of each component. Some of the large tanks were sealed without cutting and carried out of the facility by creating an opening from the building. The tanks were then transferred to a designated dismantling facility where they were shredded and stored in containers. Once the dismantling of installations and equipment is completed, contaminated concrete will be removed from the building and the designation of the controlled area will be lifted after it has been confirmed that there is no residual contamination in the building. The building will be demolished subsequently, and the site will be cleared and prepared for future use.

3.4.2.3 Reprocessing Facilities in Other Countries [14–16]

As with the dismantling of reactor facilities, the reconstruction and dismantling of reprocessing facilities are essential tasks for nuclear nations. The development of dismantling plans for reprocessing facilities or actual dismantling has already begun in the U.S. and European countries.

Reprocessing facilities currently in the dismantling process are West Valley in the U.S., the Eurochemic Reprocessing Plant in Belgium, B204 Primary Separation Plant in the U.K., AT-1 and B211 in France, and WAK in Germany. These facilities are being dismantled by applying existing techniques or techniques developed as the work proceeds, considering their individual characteristics.

For the reason that equipment is small in scale as well as from the perspective of preventing secondary contamination, conventional mechanical cutting techniques are often used in these facilities when equipment is dismantled. When dismantling buildings, one or more techniques including diamond saw, core boring, abrasive water jet and wire saw methods are chosen depending on the conditions of the buildings. For the decontamination of installations, many facilities have had system decontamination carried out following the termination of their operation, or contamination fixation undertaken to confine alpha contamination. Current technical development is primarily focused on remote dismantling techniques. At AT-1 and WAK, equipment inside cells was dismantled using hydraulic manipulators.

3.5 Site Release

Upon completing decommissioning, an application must be filed pursuant to the Reactor Regulation Act to request that the regulatory authority confirm the completion of decommissioning with regard to whether the results of the decommissioning work conform to the established standards. When the confirmation is obtained, the license for activity or installation the nuclear reactor ceases to be effective.

It is required that the application requesting the confirmation of the completion of decommissioning should contain the implementation status of the measures put forward in the decommissioning plan, and also include information on the distribution status of contamination caused by nuclear fuel materials. The criteria for confirmation are: the transfer of nuclear fuel materials has been completed; the site soil and residual facilities are in a state that they do not require measures for preventing radiation hazards; the disposal of contaminated objects has been completed; and radiation control records have been submitted to the national government.

There are several possible uses of a site (land and buildings) on which decommissioning has been completed. In the case of nuclear power plants in Japan, many of them have multiple units built on one site. Even when one of the units is decommissioned, the site may possibly continue to be used as a nuclear plant while designating the decommissioned unit and its peripheral area as a monitoring area. In the case of small-scale experimental facilities and research reactors, it is largely viewed that after a facility is dismantled, the building and land or the land can be excluded from nuclear regulatory control and put into general use without restriction.

3.5.1 Criteria for Site Release

Site release includes two concepts: unconditional release and conditional release. Unconditional release allows a site to be used freely without restriction once site release is approved. Conditional release, on the other hand, requires a facility with a certain level of residual radioactive materials to be placed under institutional control to prevent the exposure dose of individuals reusing the facility from exceeding the reference dose. Institutional control may include the designation of restricted access areas and a restriction on the duration of time spent in specified areas.

3.5.1.1 Reference Dose

The safety guide [17] issued by IAEA explains reference doses based on the following logic.

Materials once released from regulations through the clearance system, which is one example of release from regulatory control, may possibly enter into trade with a broad range of potential uses internationally and the exposure doses from such cleared items would add to the dose constraint; therefore, the reference dose is set below the order of 10 μSv/y, which is regarded as negligible in terms of the risk level. On the other hand, land (soil) and buildings continue to remain in place after their release from regulatory control; therefore, safety can be ensured as long as the exposure dose does not exceed the dose constraint of 300 μSv/y. In other words, the 300 μSv/y individual dose to a member of the general public based on the dose constraint is the starting point of the argument in this guide. It then suggests that each member country should set its own reference dose within the range of several tens to 300 μSv/y. The basis for this argument is that the dose constraint set for the boundaries of a site while the facility is in operation can be applied to the site itself upon the release of the site from regulatory control, which takes place after the operation is terminated and the facility is dismantled.

In the U.S. where many cases of site release have already occurred, the reference dose has been set at 250 μSv/y to allow some margin from the 300 μSv/y dose constraint [18]. For conditional release, the country's standard requires that the individual dose should not exceed 1 mSv/y even in the event that institutional control fails to function effectively.

In Germany, site release is positioned as part of the country's clearance system, and the reference dose is set at 10 μSv/y. The concentration of each radionuclide has been calculated based on this reference dose as explained below, and the calculated concentration values are set out in the national ordinance governing the clearance system.

3.5.1.2 Reference Radioactivity Concentration

While reference doses such as above are stipulated, confirming extremely low doses below 300 μSv/y through direct measurement is difficult. Therefore, in practice, the surface densities of the contamination of buildings and radioactivity concentrations in the soil, which correspond to the relevant reference dose, are calculated for each possible form of contamination and exposure pathway. The calculated values are then compared with values obtained through actual measurement. This process is used to judge whether site release is possible, and is the same method used to determine the clearance of wastes generated during decommissioning (see Chap. 4.2 and 8).

In the U.S., any licensee who wishes to release its site from regulatory control is required to calculate exposure doses using parameters appropriate for the characteristics of the site. It must also calculate radioactivity concentrations in the soil or surface contamination densities that correspond to the reference dose (derived concentration guideline level: DCGL) for each nuclide, and obtain approval for the calculated values through a license termination plan (LTP). The RESRAD code, which enables easy calculation of concentrations, has been developed in the U.S. It has been made widely available through the Internet and is used in different countries around the world. Germany has also used RESRAD, after making some improvements to part of the code, to calculate radionuclide concentrations based on the 10 μSv/y reference dose. Germany has set uniform reference values for radioactivity concentration and surface contamination, which have been calculated without factoring in site-specific scenarios and parameters.

When carrying out exposure dose evaluation, scenarios for the post-decommissioning use of the site need to be developed to evaluate the following: (1) the external exposure of a resident or worker or an individual temporarily entering the site to the residual radioactive material contained in the surface soil of the site; (2) their internal exposure caused by inhaling particulates; (3) exposure due to agricultural practices; and (4) internal exposure caused by intake of agricultural or livestock products cultivated or raised directly on the decommissioned site. In the case of site release, shielding material such as cover soil, which is factored in the evaluation of radioactive waste disposal (e.g., Fig. 6.3 in Chap. 6), does not exist on the site. Consequently, external exposure by direct radiation from radioactive materials in the soil contributes significantly to exposure doses. The evaluation should be made on the assumption that the agricultural and livestock products are grown directly on the decommissioned site, and radioactive materials contained in the soil transfer to the products through plant roots.

3.5.2 Verification for Site Release

In order for a site to be released from nuclear regulations, verification must be made based on the history of use of the site, measured values and other relevant data to confirm that the radioactivity concentrations or surface contamination densities of the site are kept below the values calculated through evaluation. Several countries have established verification procedures for site release, which are to be carried out in combination with necessary remedial actions on the premise that the site is contaminated with radioactive material.

The U.S. has established its radioactivity measurement and evaluation methods concerning site release by introducing statistical methods, and published a verification manual MARSSIM [19] explaining the methods. Because sites will be released from the control of nuclear regulations and put into general use, this manual was compiled with mutual consensus of multiple U.S. government agencies including the Department of Energy, Environmental Protection Agency, Department of Defense, and NRC.

As explained in Fig. 3.10, the MARSSIM approach classifies different areas of the land and buildings into those that may be affected by radioactive materials

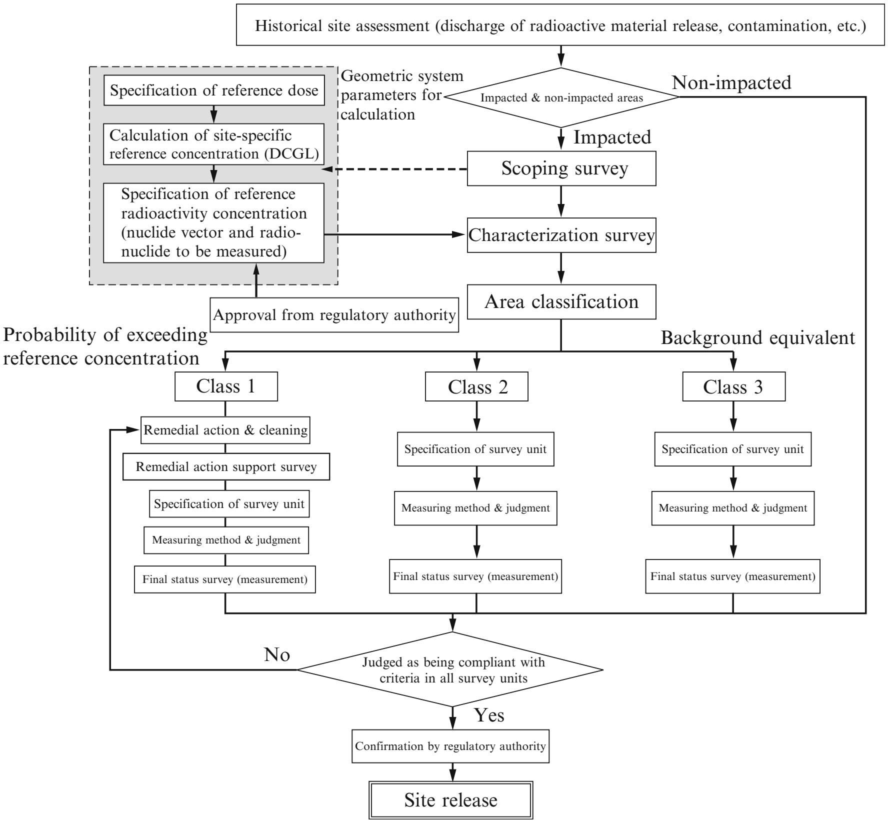


Fig. 3.10 Site release procedure in MARSSIM

(impacted areas) and those not affected by radioactive materials (non-impacted areas) based on the operating history of the site. Non-impacted areas are considered to have already met the criteria, and no measurements will be taken. For impacted areas, on the other hand, scoping and site characterization surveys are conducted followed by area classification. The percentage of the area to be covered by scans is then specified according to the area classification, and a scanning survey is conducted using a scanning instrument that has the minimum detection limit of no more than 50 or 10 % of DCGL to confirm that there are no unacceptable hot spots. Next, using a statistical method, an appropriate number of samples are taken from the site soil as well as the concrete from the building surfaces to measure their radioactivity. Upon evaluating the measurement results, the Sign test is applied when the radionuclide to be evaluated is not present in background, while the Wilcoxon Rank Sum test is applied when it is present, to determine the compliance with the criterion. The Sign test is used to test the hypothesis that the difference median is zero between the continuous distributions of two random variables. The Wilcoxon Rank Sum test is used to determine whether the distribution of the observed values of two groups is the same. For example, when ^{137}Cs is the nuclide to be evaluated, the Wilcoxon Rank Sum test is applied because ^{137}Cs is in the soil due to global fallout. In MARSSIM, Type I error or false positive, in which a null hypothesis is rejected when it is true, and Type II error or false negative, in which a null hypothesis is accepted when it is false, are each specified at 5 % in most cases.

Exercises

1. Calculate the radioactivity concentration [Bq/g] of ^{60}Co remaining in the pressure vessel material at 10 years following the termination of the operation of a reactor that had been operated for 30 effective full power years (EFPYs) at a thermal neutron flux of 1×10^{10} n(cm^2·s). The pressure vessel is made of carbon steel (density 7.9 g/cm^3), and the initial elemental ratio of ^{59}Co therein is 100 ppm. Refer to Fig. 3.4 for the thermal neutron activation cross section of ^{59}Co and the value of the half-life of ^{60}Co. Because the decrease in the number of ^{59}Co atoms due to activation is significantly smaller than the initial level, it can be assumed that the number of ^{59}Co atoms is constant.
2. Calculate the exposure dose of the general public in the vicinity via the inhalation & ingestion and surface deposition pathways when the in-air cutting of 150-m, JIS 300A × sch40 piping with a ^{60}Co surface contamination density of 1×10^4 Bq/cm^2 is performed inside a contamination control enclosure using the plasma arc technique. The calculations are to be made on the following assumptions. In order to improve the storage efficiency, the pipe is halved in the vertical direction when storing it in a 200-liter drum (diameter: 0.566 m, height: 0.8 m). Taking account of the 0.8-m drum height, the pipe is cut at intervals of 0.75 m (including the kerf width). The kerf width is 0.005 m, the contamination radioactivity dispersion rate is 30 %, no leakage from the building or contamination control enclosure is present, and the duration of exposure resulting from the dismantling work and the released radioactivity is 1 year. The collection efficiencies of HEPA or other filters of the local and building exhaust systems

are 99.0 %, respectively. The relative concentration at the site boundary is 2.2×10^{-4} (Bq/m^3)/(Bq/h), the adult breathing rate is 22.2 m^3/day, and the internal exposure dose conversion factor for ^{60}Co is 3.1×10^{-2} μSv/Bq. In addition, the surface deposition velocity is 0.01 m/s, the external exposure dose conversion factor from gamma rays associated with surface-deposited ^{60}Co is 2.2×10^{-2} (μSv/y)/(Bq/m^2), and the general public in the vicinity will be exposed, for the duration of 1 year, to surface-deposited radioactivity that has been accumulated for 1 year. It is recommended that the former Nuclear Safety Commission of Japan's Special Committee report "Evaluation of Exposure Dose of the General Public during Safety Evaluation of Light Water Nuclear Power Reactor Facilities" be used as reference when making the above calculations.

3. Calculate the minimum detectable concentration for scanning (ScanMDC) when carrying out a scanning survey of ^{60}Co on the floor surface of a turbine building. A ScanMDC can be calculated using the following formula:

$$\text{ScanMDC} = \frac{\text{MDCR}}{\sqrt{p}\,\varepsilon_i\,\varepsilon_s \dfrac{\text{Area of detector window}}{100\ \text{cm}^2}}$$

The ScanMDC is to be determined for a background level of 1,350 cpm and a 2-second interval. The performance requirements are a true positive fraction of 95 % and Type I error of 25 %. A surveyor efficiency of $p = 0.5$, instrument efficiency of $\varepsilon_i = 0.24$, and surface efficiency of $\varepsilon_s = 0.25$ are assumed. Section 6.7.2 of MARSSIM may be referred to when making calculations.

References

1. Act on the Regulation of Nuclear Source Material, Nuclear Fuel Material and Reactors (1957)
2. Nuclear Regulatory Commission, "Technology, Safety and Costs of Decommissioning a Reference Pressurized Water Reactor Power Station," NUREG/CR-0130; "Technology, Safety and Costs of Decommissioning a Reference Boiling Water Reactor Power Station," NUREG/CR-0672 (1992)
3. http://wwwndc.jaea.go.jp/jendl/j40/J40_J.html
4. ENDF/B-VII: http://www.nndc.bnl.gov/exfor/endf00.jsp
5. S. Yanagihara, T. Sukegawa, K. Shiraishi, Development of computer systems for planning and management of reactor decommissioning. J. Nucl. Sci. Technol. **38**, 193–202 (2001)
6. Nuclear Safety Commission of Japan, *Meteorological Guide for Safety Analysis of Nuclear Power Reactor Facilities* (2001)
7. International Committee on Radiation Protection, *Dose Coefficients for Intake of Radionuclides by Workers*, ICRP Publication 68, Pergamon press, Oxford (1994)
8. Y. Miyasaka, M. Watanabe, M. Tanaka, H. Nakamura, Y. Seiki, M. Tachibana, K. Ozawa, M. Hatakeyama, S. Ito, M. Yoshimori, H. Tomii, K. Nakamura, S. Yanagihara, K. Shiraishi, K. Fujiki, Results and outline of JPDR dismantling demonstration project. J. Atom. Energy Soc. Jpn. **38**, 553–576 (1996)
9. M.L. McKernan, *Shippingport Station Decommissioning Project. Final Project Report*, DOE/SSDP-0081 (1989)
10. Atomica, Dismantling of Shippingport Atomic Power Station (U.S.), (05-02-03-08)

11. Science and Technology Agency, *Decommissioning of Nuclear Facility*, (1995), p. 47
12. H. Rupar, P. Faber, M. Ruhbaum, Decommissioning of the Siemens Hanau fuel fabrication plant and hot cells. Internationale Zeitschrift fur Kernenergie **44**, 446–471 (1999)
13. S. Satomi, F. Kanayama, K. Hagiya, M. Myodo, T. Kobayashi, H. Tomii and M. Tachibana, *Removal of the Liquid Waste Storage Tank LV-2 in JRTF Part 1 (Preparatory Works)*, JAEA-Technology, 2008–067 (2008)
14. West Valley Environmental Service LLC, *West Valley Demonstration Project Progress Report*, 1 (2008)
15. X. Petitet, A. Garcia, *Decommissioning of the AT1 FBR Fuel Reprocessing Facility at La Hague*, European Commission (1998)
16. K. Heger, K.J. Birringer, H. Wiese, Lessons Learned with the Dismantling of the Karllsruhe Reprocessing Plant WAK, in *Proceedings of the International Decommissioning Symposium*, Knoxville, 12–16 June 2000
17. International Atomic Energy Agency, *Release of Sites from Regulatory Control on Termination of Practices*, IAEA Safety Guide WS-G-5.1 (2006)
18. Nuclear Regulatory Commission, 10 CFR Part 20 Subpart E(Radiological Criteria for License Termination)20.1402 (1987)
19. Four Agencies (Nuclear Regulatory Commission, Environmental Protection Agency, Department of Energy, and Department of Defense), *The Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM)*, NUREG-1575 (EPA 402-R-97-016, DOE/EH-0624) (2000) http://www.marssim.com/

Chapter 4
Clearance

Mitsuo Tachibana

4.1 What Is Clearance?

4.1.1 Necessity of a Clearance System

The term "clearance" refers to the idea that if an exposure dose due to a very low-level artificial radioactive material is sufficiently smaller than natural background radiation and human health risk is negligibly small, that artificial radioactive material does not need to be treated as a radioactive material, and therefore the material may be released from regulatory control even if the category to which the material belongs is under regulatory control for radiation protection.

Wastes generated from the dismantling of nuclear facilities may include materials to be considered for clearance (cleared items). The amount of waste generated from the decommissioning of a 1,100 MW nuclear power plant is estimated to be about 540,000 tons in the case of a boiling water reactor (BWR) and about 500,000 tons in a pressurized water reactor (PWR). The amount of cleared items generated from the decommissioning of a BWR is estimated to be 28,000 tons, which consists of 21,000 tons of metals and 7,000 tons of concrete. The amount of cleared items generated from the decommissioning of a PWR is estimated to be 12,000 tons, which includes 3,000 tons of metals and 8,000 tons of concrete. It has also been estimated that the amount of "non-radioactive wastes" (see the following

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The nuclear regulatory system in Japan has been changed significantly after the Fukushima Daiichi Nuclear Power Station accident in March 2011. Descriptions in this chapter have been translated from the book originally published in Japanese before the accident, with minimal update where appropriate. The Nuclear Regulation Authority newly established after the accident has not completed its review for the guidelines and regulations established by the former Nuclear Safety Commission. In this chapter, guidelines set by the NSC have been adopted.

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section) generated from a BWR is 495,000 tons and that generated from a PWR is 477,000 tons. In contrast, the amount of cleared items generated from 20 years of operation of a PWR is estimated to be 650 tons, which consists of 500 tons of metals and 150 tons of concrete. Thus, the amount of cleared items generated from operation of a nuclear power plant is only about 1/20 of that of cleared items generated at the time of decommissioning. The amount of cleared items generated from decommissioning, therefore, is by far greater.

A clearance level for a radioactive material is the level at or below which the special nature of the material as a radioactive material does not need to be considered. The establishment of the clearance system has made it possible to increase the number of options for rational recycling, reuse or disposal of a large amount of wastes generated from R&D and power generation uses of nuclear energy. In other words, under the clearance system, environmental loading can be reduced by utilizing recyclable or reusable wastes generated from controlled areas of nuclear facilities. Wastes that cannot be recycled or reused with reasonable effort may be buried as a means of disposal. It can be said that the clearance system contributes to the creation of a zero-waste society, which is the goal Japan is trying to achieve.

Since the materials and objects that are recycled or reused under the clearance system can be distributed in any part of society, clearance levels need to be consistent in the international community. Efforts to determine clearance levels have been underway, therefore, under the leadership of the International Atomic Energy Agency (IAEA) and the European Commission (EC). Details of the studies by these international organizations are described in Sect. 4.5.

4.1.2 Concepts Similar to Clearance

“Exemption” and “exclusion” are concepts related to regulatory control associated with radiation protection and similar to the concept of clearance.

Exemption means that an artificial radioactive material does not need to be placed under regulatory control for radiation protection because the human health impact of the artificial radioactive material is so small as to be negligible. Materials and objects in very small quantities such as radioactive tracers used in research and calibration sources as well as some consumer goods such as incandescent gas mantles, optical lenses and ceramic tableware containing very few nuclides fall into this category. Exposure doses due to these artificial radioactive materials are sufficiently smaller than natural background radiation. Radioactivity concentrations of nuclides used as criteria for exemption are termed exemption levels.

Exclusion means the deliberate exclusion of a naturally occurring radioactive material (NORM) from the scope of regulatory control because exposure to that material cannot be controlled and therefore regulatory control is not possible or because regulatory control is expected to have little effect. Naturally occurring nuclides include nuclides that are as old as the earth and nuclides that are naturally produced by cosmic rays. Nuclides falling into the former category include ^{40}K, ^{87}Rb, ^{138}La, ^{147}Sm, ^{176}Lu and the ^{232}Th series and ^{238}U series nuclides. Nuclides produced by cosmic rays include ^{3}H, ^{7}Be, ^{14}C, ^{22}Na and ^{36}Cl. These nuclides are present in the soil and rock and in the air.

A NORM whose radioactivity concentration has been artificially enhanced is called a TENORM (technologically enhanced NORM). TENORMs include monazite used as raw materials for paints and hot spa elements, phosphate ore used to manufacture ammonium phosphates, and titanium ore used to manufacture titanium oxides. The average exposure dose of workers handling these types of ores is as high as 0.3 mSv/y for monazite, 0.28 mSv/y for phosphate ore, and 0.27 mSv/y for titanium ore.

Besides the three concepts described above, there is also the concept of "non-radioactive wastes." This term means wastes that are generated in a controlled area but are deemed not to be radioactive wastes in view of their history of use. Thus, the term refers to wastes that cannot cause contamination by radioactive materials or that do not require consideration of the effect of activation by neutron rays. This concept has been applied to the dismantling of the Japan Power Demonstration Reactor (JPDR) and the replacement of steam generators of commercial nuclear power plants.

The Radioactive Waste Safety Subcommittee in the Nuclear and Industrial Safety Subcommittee, Advisory Committee for Natural Resources and Energy, Ministry of Economy, Trade and Industry (METI), deliberated on the concept of "non-radioactive wastes," contamination prevention measures, detection limits, and the methodology for selecting parts and components classified as "non-radioactive wastes." In October, 2007, the results of the deliberations were documented into a report on practical criteria for the handling of non-radioactive wastes [1].

4.1.3 The Japanese Clearance System

In Japan, clearance levels were first derived by the former Nuclear Safety Commission (NSC) of Japan. On the basis of the results thus obtained, the regulatory authorities, namely, the former Nuclear and Industrial Safety Agency (NISA) of METI and the Ministry of Education, Culture, Sports, Science and Technology (MEXT), deliberated on the introduction of a clearance system for reactor facilities and nuclear fuel use facilities.

In 1997 the NSC of Japan began to consider clearance levels from a scientific viewpoint. On the basis of the clearance level derivation approach indicated in IAEA's technical document "Clearance Levels for Radionuclides in Solid Materials: Application of Exemption Principles," TECDOC-855 [2], three committee reports [3–5] were prepared for application to concrete and metals generated from the operation and decommissioning of (1) light water reactors and gas-cooled reactors used for power generation and research reactors, (2) heavy water reactors and fast reactors used for research, and (3) nuclear fuel use facilities.

In order to determine clearance levels, it is necessary to identify nuclides to be considered (termed "nuclides to be studied") and assume evaluation pathways for exposure dose derivation. In the committee reports [3–5], a total of 58 nuclides were selected as those contained in wastes generated from the dismantling of reactor facilities and nuclear fuel use facilities, having a great impact on the human body and existing in relatively large quantities in such wastes. The term "nuclide to be studied" here then specifically means a nuclide for which clearance levels are to be derived.

Evaluation pathways for exposure dose derivation are categorized into two groups: evaluation pathways associated with recycling or reuse and evaluation pathways associated with burial disposal of wastes. In view of these categories of evaluation pathways that are likely to occur in reality, a total of 77 evaluation pathways were selected for recycling or reuse, and 125 evaluation pathways were selected for burial disposal. From these pathways, evaluation pathways that are likely to cause exposure doses much lower than those caused by other evaluation pathways and evaluation pathways that are deemed to be included in other evaluation pathways were excluded. As a result, 32 evaluation pathways for recycling or reuse and 41 evaluation pathways for burial disposal were selected.

For nuclear fuel use facilities, evaluation pathways whose impact is obviously small judging from the clearance level derivation results for reactor facilities were excluded. On the other hand, recycling for use in lead storage batteries for automobiles was added as a lead-related evaluation pathway because wastes generated from the dismantling of nuclear fuel use facilities contain a large amount of lead used as shielding materials.

In the BSS (Basic Safety Standards) [6], IAEA indicated the concepts of exclusion, exemption and clearance, along with the exemption levels for quantities of the order of 1 ton. Later in 2004, IAEA indicated in the IAEA Safety Guide RS-G-1.7, "Application of the Concepts of Exclusion, Exemption and Clearance" [7], radioactivity concentrations at which exclusion or exemption may be applied to large quantities of materials for each naturally occurring nuclide and artificial nuclide. The IAEA Safety Guide RS-G-1.7 states that these radioactivity concentrations can be used as the basis for clearance.

In view of these IAEA guidelines, the NSC of Japan reviewed the previously derived clearance levels. The nuclides for which the derived values before and after the review differed by a factor of 10 or more were ^{59}Ni and ^{63}Ni, and for most of the nuclides there were no significant differences. The NSC of Japan also deliberated on the clearance level verification method by which to check whether the values under consideration are at or below the clearance levels. In 2001, a report [8] was prepared on this.

In response to the clearance level derivation results indicated by the NSC of Japan [9], the clearance levels and the clearance level verification methods for reactor facilities and nuclear fuel use facilities were evaluated by the Radioactive Waste Safety Subcommittee in the Nuclear and Industrial Safety Subcommittee, Advisory Committee for Natural Resources and Energy, METI, and the Reactor Safety Regulation Committee of the Science and Technology Policy Bureau, MEXT, respectively [10, 11]. As a result, the clearance system for reactor facilities and nuclear fuel use facilities was established under the Act on the Regulation of Nuclear Source Material, Nuclear Fuel Material and Reactors (Reactor Regulation Act).

4.2 Methods of Clearance Level Derivation

When deriving clearance levels, it is common practice to follow the procedure described below on the basis of the reference dose values for radiation protection, which are said to be an approximate guide to exposure doses.

For clearance application, the reference dose values are specified first, and evaluation items are selected. Then, evaluation pathways for exposure dose evaluation and persons to be evaluated are selected, and an evaluation model used to derive the exposure dose in each evaluation pathway is constructed. Scenario parameters are then defined, and the radioactivity concentration of each of the nuclides to be studied corresponding to the reference dose values is derived. In the next step, the derivation results for different evaluation pathways are put together for each nuclide to be studied, and the evaluation pathway indicating the lowest radioactivity concentration is taken as the derived value of the clearance level for the nuclide to be studied under consideration. This evaluation pathway is used as the basis for clearance level derivation for the nuclide to be studied under consideration and is therefore called a “determined pathway.”

This section deals with methods of clearance level derivation for reactor facilities and nuclear fuel use facilities. Detailed methods of clearance level derivation are shown in Chap. 8.

4.2.1 Preconditions for Clearance Level Derivation

4.2.1.1 Reference Dose Values

In order to derive a clearance level, it is necessary to set a standard by which to specify the extent of allowable human exposure, and that standard is termed a “reference dose value.” With respect to a basic approach to reference dose values’ determination and indices used for that purpose, international organizations such as the Food and Agriculture Organization (FAO), the International Labour Organization (ILO) and the World Health Organization (WHO) of the United Nations unanimously have a criterion that “risk to human health is negligible,” [6] and this opinion is shared by the IAEA, too.

The levels of negligible risk are generally thought to be 10^{-6} to 10^{-7}/y [12, 13], which translate to exposure doses of 10–100 μSv/y [12]. These values not only are sufficiently smaller than the individual dose limit for people living in the vicinity of a nuclear facility as defined by law (1 mSv/y) and the average exposure dose from natural background radiation (2.4 mSv/y) but also are about the same as the range of variation of 2.4 mSv/y (one to several percent, or 20–100 μSv/y). Judging from these, the level of negligible risk to human health is a few tens of μSv/y. In view of the possibility of exposure to multiple sources, it is thought that the individual effective dose to be used as the reference dose values for the clearance level derivation should be 10 μSv/y. The equivalent dose at the skin is 50 mSv/y.

4.2.1.2 Evaluation Items

The operation and decommissioning of reactor facilities, such as light water reactors, gas-cooled reactors, heavy water reactors and fast reactors, as well as nuclear fuel use facilities that handle fuel and materials, such as post-irradiation examination facilities and research facilities, generate solid materials containing radioactive materials. Among the solid materials thus generated, metals and concrete that are thought to meet the clearance level requirements and are generated in large quantities are to be evaluated. Gaseous and liquid wastes are not thought of as evaluation items because standard values have been specified separately for exhaust air and waste water management.

The exemption levels in the IAEA guidelines are not intended for application only to the decommissioning of reactor facilities. Instead, they are also intended for application to nuclear fuel use facilities other than reactor facilities. Since, however, it has been found that the quantity of wastes generated by the dismantling of reactor facilities is by far greater, it has been decided that the quantity of evaluation items is 500,000 tons of wastes generated from the dismantling of a 1,100 MW nuclear power plant, which consists of 50,000 tons of cleared items and 450,000 tons of non-radioactive wastes.

The weight ratio of the cleared metals in the metals to be recycled is 0.1, and the weight ratio of the cleared concrete in the concrete to be recycled is 0.1. The mixing ratio of evaluation items and cleared items is termed the weight ratio of the cleared items in the wastes, and its value is, in this case, 0.1.

Clearance levels are evaluated in terms of radioactivity concentration averaged for a unit quantity of at least about 10 tons. Variability of the distribution of radioactivity concentration in the cleared item is permitted to a certain degree.

As mentioned in Sect. 4.1.3, a total of 58 nuclides have been specified as nuclides to be studied that are contained in the wastes generated by the dismantling of reactor facilities and nuclear fuel use facilities, have a significant impact on the human body and occur in relatively large quantities of the wastes.

4.2.1.3 Evaluation Pathways

Cleared items are specified on the condition that cleared items are allowed to be used for any purpose and to be taken anywhere on the assumption that in the evaluation pathways associated with recycling, metals are used in consumer goods or as construction materials and concrete is used as construction materials. In the evaluation pathways associated with burial disposal, it is assumed that concrete and metals are buried at a waste disposal facility in the same manner as industrial wastes.

The evaluation pathways considered for the clearance level derivation are selected taking into consideration the possibility of exposure of workers and local residents while evaluation items are recycled or reused or buried for disposal.

Although many exposure cases due to evaluation items are expected, it is common practice to select exposure cases that involve high exposure doses and that are expected to occur with a high frequency as evaluation pathways.

Evaluation pathways for which exposure dose evaluation is to be made for the clearance level derivation have been determined by first listing all realistically possible evaluation pathways associated with cleared items, and excluding evaluation pathways that are thought to involve low exposure doses and evaluation pathways included in other evaluation pathways. As a result, 32 evaluation pathways were selected for recycling and reuse of metals and concrete and 41 evaluation pathways have been selected for burial disposal. Figure 4.1 shows the recycling and reuse-related evaluation pathways used for the clearance level derivation, and Fig. 4.2 shows the evaluation pathways associated with burial disposal.

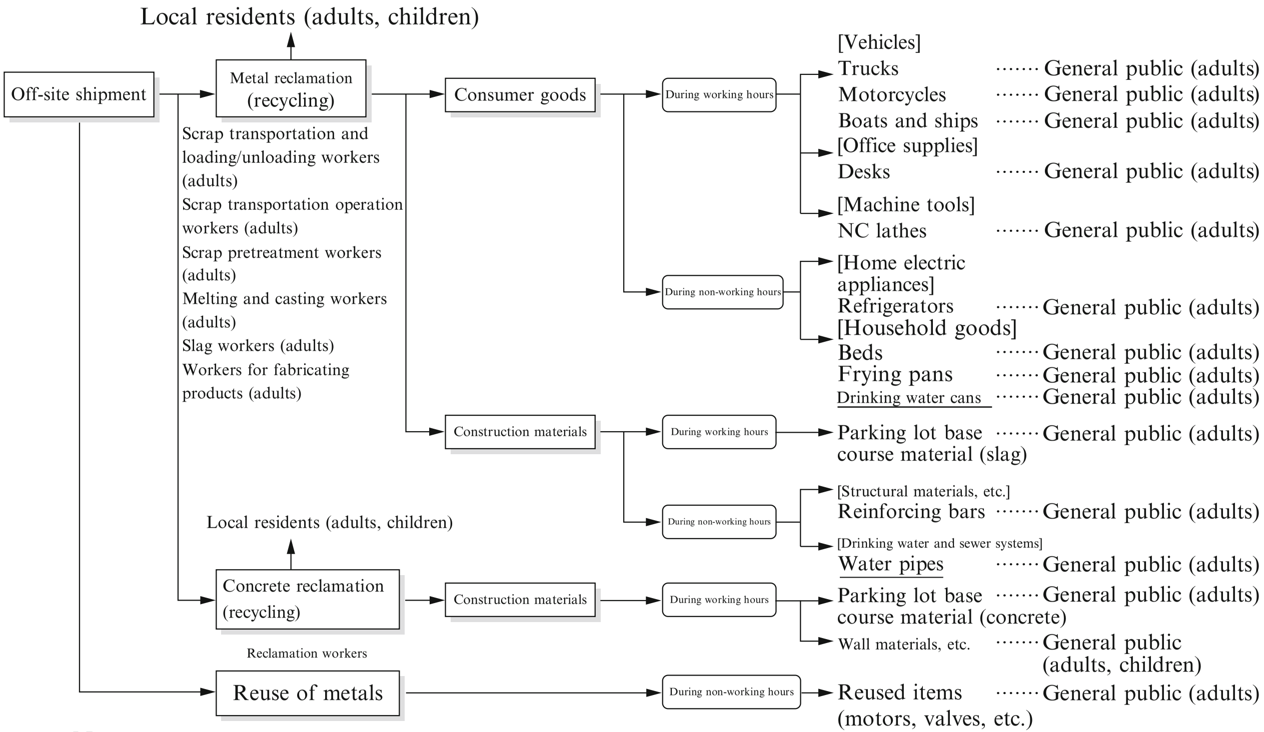
For nuclear fuel use facilities, recycling and reuse-related evaluation pathways for lead were added because shielding materials used at nuclear fuel use facilities contain large quantities of lead.

The next step is to select persons to be evaluated. In this step, one person representative of a representative homogeneous group that is likely to be exposed to the highest dose in a realistically possible evaluation pathway is selected for the clearance level derivation. Persons to be evaluated include adult workers who directly work on or with cleared items, adults engaged in agriculture, adult residents and 1- or 2-year-old child residents. For groups of these types of people, external exposure, inhalation intake exposure, ingestion intake exposure, and skin exposure doses along each evaluation pathway are derived.

4.2.2 Exposure Dose Evaluation Models

In the preceding sections, reference dose values, evaluation items and evaluation pathways have been determined. This section deals with mathematical models for deriving external exposure doses, inhalation intake exposure doses, ingestion intake exposure doses and skin exposure doses along the evaluation pathways shown in Figs. 4.1 and 4.2.

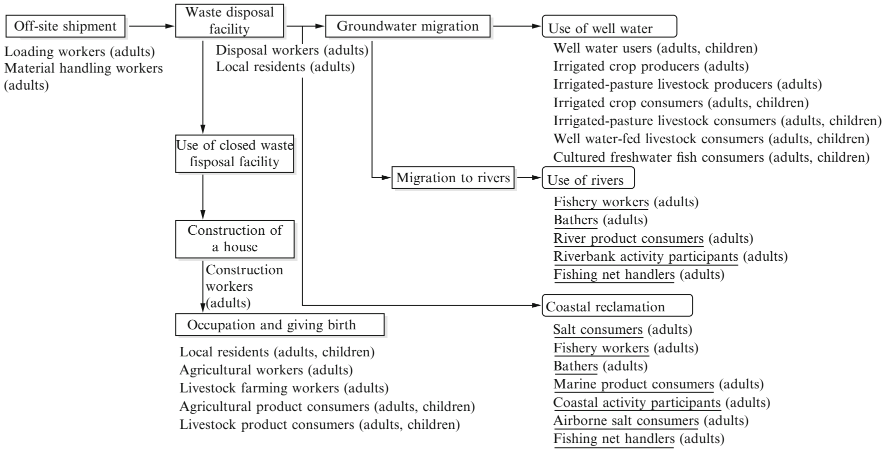
In the clearance level derivation, various scenario parameters are used depending on the evaluation pathways involved. Many of these scenario parameters are not single constant values; instead, they vary with certain patterns and certain widths. In view of the social environment and life styles in Japan, therefore, parameter values that are thought to be realistic and average are selected from the values found in literature. For scenario parameters for which realistic values are difficult to determine because directly relevant literature and sufficient amounts of data are not available, in many cases other parameter values are used as a guide or conservative values within a practical range are used. The parameter values used in the four evaluation models are shown in Table 8.1 of Chap. 8, “Derivation of Clearance Levels”.



Notes

1. Underlined items are in an evaluation pathway that is excluded in clearance level derivation for nuclear fule use facilities.
2. The recycling of lead (for lead storage batteries for automobiles) is evaluated in the clearance level derivation for nuclear fuel use facilities.

Fig. 4.1 Evaluation pathways associated with recycling or reuse for clearance level derivation



Note : Underlined items are in an evaluation pathway that is excluded in clearence level derivation for nuclear fuel use facilities

Fig. 4.2 Evaluation pathways associated with burial disposal for clearance level derivation

4.2.2.1 External Exposure Dose Evaluation Model

If the radioactivity concentration of nuclide i in a recycled product is represented by $C_m(i)$ [Bq/g], the external exposure dose $D_{EXT}(i)$ [Sv/y] due to cleared items or recycled products such as consumer goods and construction materials produced by recycling cleared items can be derived as Eq. (4.1).

$$D_{EXT}(i) = C_m(i) \cdot S \cdot t_f \cdot \frac{1 - \exp(-\lambda_i t_i)}{\lambda_i t_i} \cdot D_{CF,EA}(i) \tag{4.1}$$

S: shielding factor for external radiation,
t_f: annual exposure time during the exposure scenario,
λ_i: decay constant for nuclide i [y^{-1}],
t_i: exposure time during the exposure scenario [y], and
$D_{CF,EA}(i)$: dose conversion factor for external exposure to nuclide i [(Sv/y)/(Bq/g)].

The annual exposure time during the exposure scenario t_f used here is the ratio of time during which a recycled product is used in a year. The shielding factor S for external radiation has a value of 1 if the person to be evaluated is not shielded by a wall-like barrier during the service life of the recycled product. If external exposure continues for a long period of time, it is necessary to take into account a decay in radioactivity concentration during the period of external exposure to nuclide i.

External exposure due to the surface contamination of a reused item, which is a cleared item used as-is, is derived according to Eq. (4.1). In this derivation, the radioactivity concentration $C_m(i)$ of nuclide i in the recycled product shown in Eq. (4.1) is replaced with surface contamination density $C_s(i)$ [Bq/m^2] of nuclide i of the reused item, and the dose conversion factor for external exposure per unit radioactivity concentration, $D_{CF,EA}(i)$, of nuclide i is replaced with dose conversion factor for external exposure per unit surface contamination density, $D_{CF,ES}(i)$ [Bq/m^2].

The dose conversion factor for external exposure for each evaluation pathway is derived for different recycled products or reused items as radiation sources by using a shielding calculation code based on the point kernel integration method and the one-dimensional neutron transport on Sn method. In these derivation processes, the properties and geometries of recycled products are approximated as homogeneous volume sources, infinite plate sources or surface contamination sources, and self-absorption is taken into consideration in modeling.

The radioactivity concentration of recycled products requires different methods of derivation for different evaluation pathways. This is because it is necessary to reflect differences in the type of work such as scrap transportation or landfill, differences in materials such as construction materials (e.g., wall materials) produced by using the recycled coarse aggregate or metal products made of the recycled metal, and differences in the place of exposure such as parking lot base course materials or wall materials produced by using the recycled coarse aggregate.

For example, as shown in Fig. 4.3, the radioactivity concentration $C_m(i)$ of nuclide i of a metal product made by recycling cleared metals is derived as Eq. (4.2).

$$C_m(i) = C_{WM}(i) \cdot F_{MC} \cdot T_i(i) \cdot G_M \cdot \exp\left(-\lambda_i t_{pd}\right) \tag{4.2}$$

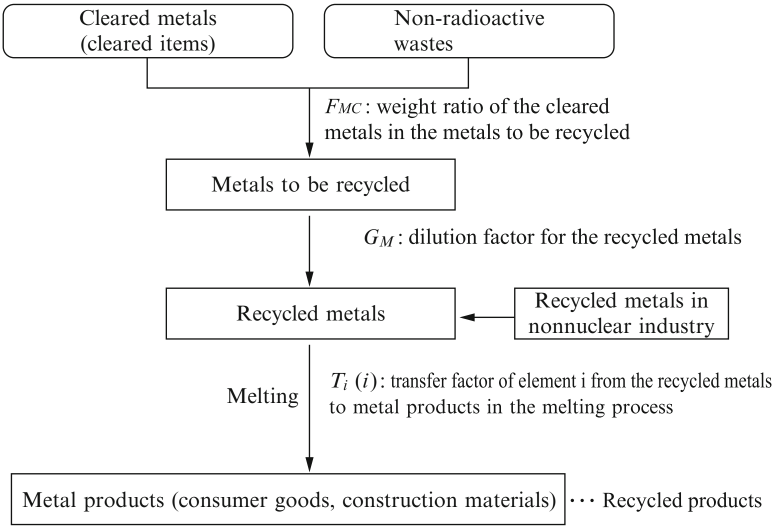


Fig. 4.3 Flow of main recycling processes of cleared metals

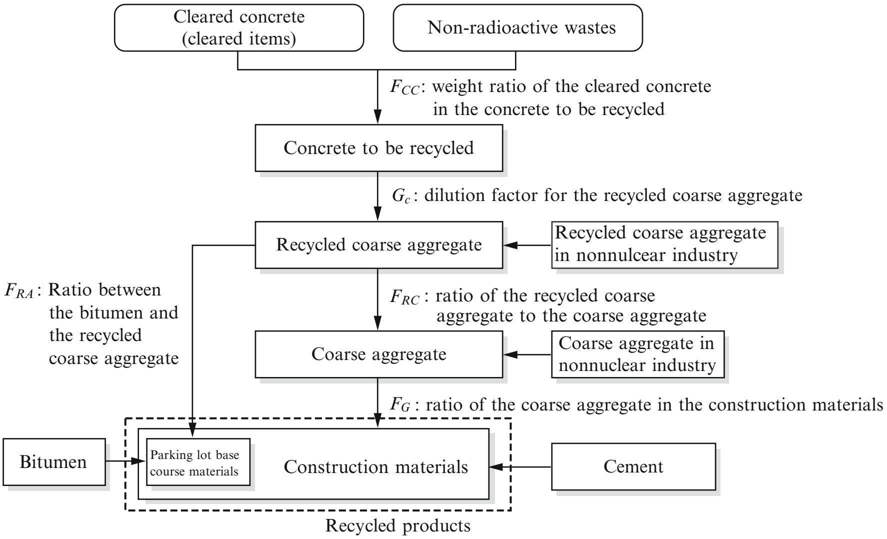


Fig. 4.4 Flow of main recycling processes of cleared concrete

$C_{WM}(i)$: radioactivity concentration of nuclide i in the cleared metals [Bq/g],
F_{MC}: weight ratio of the cleared metals in the metals to be recycled,
$T_i(i)$: transfer factor of element i from the recycled metals to the metal products in the melting process,
G_M: dilution factor for the recycled metals, and
t_{pd}: duration from clearance to recycling [y].

The dilution factor G_M for the recycled metals is ratio of the quantity of the recycled metals containing nuclides to the total quantity of recycled metals in cases where recycled metal containing nuclides is mixed with recycled metals in the nonnuclear industry that do not contain nuclides.

As shown in Fig. 4.4, the radioactivity concentration $C_m(i)$ of nuclide i contained in the parking lot base course material produced by using the recycled coarse aggregate made of cleared concrete is derived as Eq. (4.3).

$$C_m(i) = C_{WC}(i) \cdot F_{CC} \cdot G_C \cdot F_{RA} \cdot \exp\left(-\lambda_i t_{pd}\right) \tag{4.3}$$

$C_{WC}(i)$: radioactivity concentration of nuclide i in the cleared concrete [Bq/g],
F_{CC}: weight ratio of the cleared concrete in the concrete to be recycled,
G_C: dilution factor for the recycled coarse aggregate, and
F_{RA}: ratio between the bitumen and the recycled coarse aggregate.

4.2.2.2 Inhalation Intake Exposure Dose Evaluation Model

Internal exposure, which occurs when local residents or workers inhale dust from recycled, reused or buried cleared items or dust generated by objects contaminated by cleared items, is evaluated by using the mass concentration method or the resuspension factor method.

In the mass concentration method, the radioactivity concentration of dust in the air inhaled is given as a direct scenario parameter, and internal exposure doses are derived from the breathing rate of the person to be evaluated. In the resuspension factor method, internal exposure doses are determined by deriving the radioactivity concentration of dust in the air from the surface contamination density of cleared items, the ratio of resuspendable loose contamination, and the resuspension factor.

The radioactivity concentration of dust in the air generated by the melting of scrap metal is derived by taking into account the transfer factor of elements into the air during the melting process and the apparent concentration factor.

4.2.2.3 Ingestion Intake Exposure Dose Evaluation Model

For ingestion intake exposure, three types of ingestion intake are assumed: (1) ingestion intake of loose contaminants from reused items; (2) ingestion intake of radioactive materials leached out of recycled metal products; and (3) ingestion intake of well water and agricultural, livestock and aquatic products containing nuclides.

Exposure due to ingestion intake of loose contaminants is thought to occur when a worker uses a reused item and loose contaminants are transferred to the hands of the worker so that the contaminants are unintentionally taken into the body. Exposure doses are derived from the surface contamination density of the reused item, the ratio of resuspendable loose contamination, and ratio of ingestion intake of loose contamination. Exposure due to dust generated by resuspension of loose contaminants is derived by using the inhalation intake exposure dose evaluation model described in Sect. 4.2.2.2.

It is assumed that exposure due to the ingestion intake of radioactive materials leached out of recycled metal products results from the ingestion intake of radioactive materials leached out of metal products produced by melting and fabricating cleared items. In this case, the internal exposure dose D_{ING} (i) is derived from Eq. (4.4).

$$D_{ING}(i) = C_m(i) \cdot A_f \cdot t_f \cdot R_e \cdot \rho_e \cdot \frac{1 - \exp(-\lambda_i t_i)}{\lambda_i t_i} \cdot D_{CF,ING}(i) \tag{4.4}$$

$C_m(i)$: radioactivity concentration of nuclide i in the metal product [Bq/g],
A_f: surface area of the metal product [m^2],
t_f: annual exposure time during the exposure scenario,
R_e: metal corrosion rate [m/y],
ρ_e: density of the metal [g/m^3], and
$D_{CF,ING}$ (i): dose conversion factor for ingestion intake of nuclide i [Sv/Bq].

The annual exposure time during the exposure scenario t_f is ratio of the time during which the metal product is used in a year.

The third type of exposure, that is, exposure due to the ingestion intake of well water or agricultural, livestock or aquatic products containing nuclides, is an evaluation pathway that assumes that wastes as a mixture of cleared items and "non-radioactive wastes" have been buried as industrial wastes, and some of the nuclides contained in the wastes have leached from them. Exposure is evaluated, therefore, for a case where the nuclides that leaked from the waste disposal facility have entered the well water through groundwater, and some of the well water has been drunk; and exposure doses are derived for a case where agricultural crops produced by using that well water for irrigation have been eaten.

Here, exposure dose is evaluated by assuming that the wastes were buried at the waste disposal facility, and some of the nuclides contained in the wastes leach into rainwater that has infiltrated the waste disposal facility and then they enters groundwater. The leakage rate of nuclide from the waste disposal facility is derived by using the emission coefficient model [14] proposed by the UK Radiation Protection Agency, which uses data on the radioactivity concentrations of nuclides in the wastes, the leakage rate of nuclide and the weight of the wastes. The nuclides that have leaked from the waste disposal facility enters an aquifer and migrates through the aquifer together with groundwater. The radioactivity concentration of nuclides in groundwater is derived by assuming that the density and porosity (void ratio) of the aquifer soil and the thickness of the aquifer are constant and by applying an advection-dispersion equation that allows for the retardation of nuclide migration due to sorption to soil, dispersion of groundwater flow, direction and groundwater flows. The analysis method is described in detail in Sect. 8.4.3.3.

The nuclide concentration of well water can be derived by taking into account the distance from the point of inflow into the aquifer from the waste disposal facility to the well and the mixing, or dilution, between the groundwater containing the nuclides and the surrounding groundwater that does not contain them. By using the annual drinking water intake consumed per year, the exposure dose due to the ingestion intake of well water can be determined.

If well water is used to irrigate agricultural land, it is assumed that the nuclides contained in the well water are accumulated in the soil. The radioactivity concentration of the nuclides in the irrigation water equals that in the well water, and the radioactivity concentration of the nuclides in the irrigation water can be derived by taking into account the ratio of the quantity of the nuclides left in the soil to the quantity of the nuclides contained in the irrigation water applied. By using annual intake rate for agricultural crops, the exposure dose due to the ingestion intake of agricultural crops can be determined.

4.2.2.4 Skin Exposure Dose Evaluation Model

Skin exposure that occurs when a worker touches a cleared item and a certain amount of nuclides is transferred to the surface of the body such as the hands of the worker. This kind of exposure is evaluated by taking into account the dilution factor for the cleared item and the concentration factor for dust, and using the layer thickness and the density of dust loading on the skin. In the evaluation of skin exposure, it is necessary to take into consideration exposure to beta rays and gamma rays from the nuclides attached to the body surface. Since beta rays and gamma rays differ in penetrating power, there is a difference in the thickness of the radiation-sensitive layer of the skin corresponding to the dose conversion factor. The dose conversion factors for the radiation-sensitive layer corresponding to 4 and 7 mg/cm^2 are used for beta rays and gamma rays, respectively.

4.2.3 Clearance Levels

4.2.3.1 Clearance Levels in Japan

The former NSC of Japan has indicated the clearance levels of 58 nuclides for reactor facilities and nuclear fuel use facilities, while the IAEA Safety Guide RS-G-1.7 indicates exemption levels, instead of clearance levels, for 277 artificial nuclides. The IAEA Safety Guide RS-G-1.7 states that the exemption levels can be used as a basis for clearance level application.

Main sources of contamination due to wastes generated from the decommissioning of reactor facilities are secondary contamination due to adhesion of leaked reactor coolant to surfaces of equipment and structures and infiltration of leaked reactor coolant into building structure and activated contamination due to the effect of neutron rays in the reactor. The main source of contamination due to wastes generated by the decommissioning of nuclear fuel use facilities is secondary contamination due to adhesion of chips and fines produced during the cutting and grinding of irradiated samples to surfaces of equipment and structures. Thus, contamination characteristics of wastes generated from decommissioning vary among different types of facilities. By referring to the results of clearance level deliberations made by the NSC of Japan and the IAEA Safety Guide RS-G-1.7, therefore, a METI ordinance stipulates clearance levels of 33 nuclides applicable to power generation reactors, and a MEXT ordinance stipulates clearance levels of 33 nuclides for research reactors such as light water reactors, heavy water reactors and fast reactors and 49 nuclides for facilities handling irradiated fuel and materials classified as nuclear fuel use facilities.

Table 4.1 shows the clearance levels of different nuclides specified by the NSC of Japan, IAEA, the METI ordinance and the MEXT ordinance.

Table 4.1 Clearance levels (Bq/g)

Nuclide	NSC of Japan	IAEA (RS-G-1.7)	METI Ordinance[a]	MEXT Ordinance[b]	
	Clearance level	Exemption level	Power generation reactors	Reactor facilities	Nuclear fuel use facilities
^{3}H	60	100	100	100	100
^{14}C	4	1	1	1	1
^{36}Cl	0.3	1	1	1	–
^{41}Ca	100	–	100	100	–
^{46}Sc	1	0.1	0.1	0.1	0.1
^{54}Mn	2	0.1	0.1	0.1	0.1
^{55}Fe	2,000	1,000	1,000	1,000	1,000
^{59}Fe	4	1	1	1	1
^{58}Co	3	1	1	1	1
^{60}Co	0.3	0.1	0.1	0.1	0.1
^{59}Ni	30	100	100	100	–
^{63}Ni	100	100	100	100	–
^{65}Zn	2	0.1	0.1	0.1	0.1
^{89}Sr	200	1,000	–	–	1,000
^{90}Sr	0.7	1	1	1	1
^{91}Y	400	100	–	–	100
^{95}Zr	2	1	–	–	1
^{94}Nb	0.2	0.1	0.1	0.1	0.1
^{95}Nb	8	1	1	1	1
^{99}Tc	1	1	1	1	–
^{103}Ru	10	1	–	–	1
^{106}Ru	6	0.1	0.1	0.1	0.1
^{108m}Ag	0.2	–	0.1	0.1	0.1
^{110m}Ag	0.6	0.1	0.1	0.1	0.1
^{114m}In	50	10	–	–	10
^{113}Sn	8	1	–	–	1
^{119m}Sn	1,000	–	–	–	1,000
^{123}Sn	300	–	–	–	300
^{124}Sb	2	1	1	1	1
^{125}Sb	2	0.1	–	–	0.1
^{123m}Te	9	1	1	1	–
^{125m}Te	50	1,000	–	–	1,000
^{127m}Te	20	10	–	–	10
^{129m}Te	20	10	–	–	10
^{129}I	0.5	0.01	0.01	0.01	–
^{134}Cs	0.5	0.1	0.1	0.1	0.1
^{137}Cs	0.8	0.1	0.1	0.1	0.1
^{133}Ba	2	0.1	0.1	0.1	–
^{141}Ce	80	100	–	–	100

(continued)

Table 4.1 (continued)

Nuclide	NSC of Japan	IAEA (RS-G-1.7)	METI Ordinance[a]	MEXT Ordinance[b]	
	Clearance level	Exemption level	Power generation reactors	Reactor facilities	Nuclear fuel use facilities
^{144}Ce	30	10	–	–	10
^{148}Pm	3	–	–	–	3
^{152}Eu	0.4	0.1	0.1	0.1	–
^{154}Eu	0.4	0.1	0.1	0.1	0.1
^{155}Eu	10	1	–	–	1
^{153}Gd	20	10	–	–	10
^{160}Tb	3	1	1	1	1
^{181}Hf	8	1	–	–	1
^{182}Ta	2	0.1	0.1	0.1	0.1
^{238}Pu	0.2	0.1	–	–	0.1
^{239}Pu	0.2	0.1	0.1	0.1	0.1
^{240}Pu	0.2	0.1	–	–	0.1
^{241}Pu	10	10	10	10	10
^{241}Am	0.2	0.1	0.1	0.1	0.1
^{242m}Am	0.2	0.1	–	–	0.1
^{243}Am	0.2	0.1	–	–	0.1
^{242}Cm	3	10	–	–	10
^{243}Cm	0.3	1	–	–	1
^{244}Cm	0.4	1	–	–	1

[a] METI Ordinance No. 112: Rules for Verification of the Concentration of Radioactive Materials Contained in Goods Used in Refinery Plants and Other Facilities (November 22, 2005)

[b] MEXT Ordinance No. 49: Rules for Verification of Concentration of Radioactivity Related to Reactors Used for Research (November 30, 2005)

4.2.3.2 Clearance Levels in Other Countries

In other countries such as Germany and Sweden, various clearance systems have been established, and recycling or burial disposal of cleared items is being practiced under those systems.

In Germany, clearance levels have been stipulated according to the type of waste and various conditions. The criteria for unconditional clearance for ^{60}Co are 0.1 Bq/g for solid and liquid materials, 0.03 Bq/g for surface soil, and 0.09 Bq/g for building rubble and excavated soil with amount higher than 1,000 tons per year. The criteria for conditional clearance are 4 Bq/g for solid and liquid materials to be disposed of and 0.6 Bq/g for scraps to be recycled. Besides clearance levels for nuclide concentration, clearance levels for surface contamination density are also stipulated. For ^{60}Co, the criterion for unconditional clearance for buildings to be reused is 0.4 Bq/cm^2, and the criterion for conditional clearance for buildings to be pulled down is 3 Bq/cm^2.

In Sweden, the clearance level for metals to be recycled is 0.5 Bq/g, and that for nuclides emitting alpha rays is 0.1 Bq/g. In terms of surface contamination density, the clearance level for nuclides emitting alpha rays is 0.4 Bq/cm^2, and that for nuclides emitting beta rays and gamma rays is 4 Bq/cm^2. It has also been stipulated that the clearance level for nuclides to be buried at industrial disposal facility are 5 Bq/g for nuclides emitting beta rays and gamma rays and 0.5 Bq/g for nuclides emitting alpha rays. It is additionally required that the total amount of radioactivity per site be limited to 1 GBq/y.

These clearance levels have been determined mainly as a result of historical circumstances or according to the detection limits of radiation measuring instruments. Consequently, they are not necessarily based on radiological effects, nor are they internationally consistent. Because cleared scraps are likely to be distributed internationally, clearance levels should be internationally standardized.

4.3 Clearance Level Verification

4.3.1 Flow of Clearance Level Verification

Clearance level verification is the process in which a nuclear operator measures a cleared item, evaluates it to verify, by using a clearance level, that it is an item that does not need to be treated as a radioactive material, and ensures, with an appropriate involvement of the regulatory authority, that the evaluation is correct. As shown in Fig. 4.5, nuclear operators are required to perform a clearance level verification at five stages, namely: (1) preliminary investigation; (2) selection of cleared items; (3) determination of radioactivity concentration measurement and judgment methods; (4) measurement and judgment of cleared items; and (5) storage, management and off-site shipment.

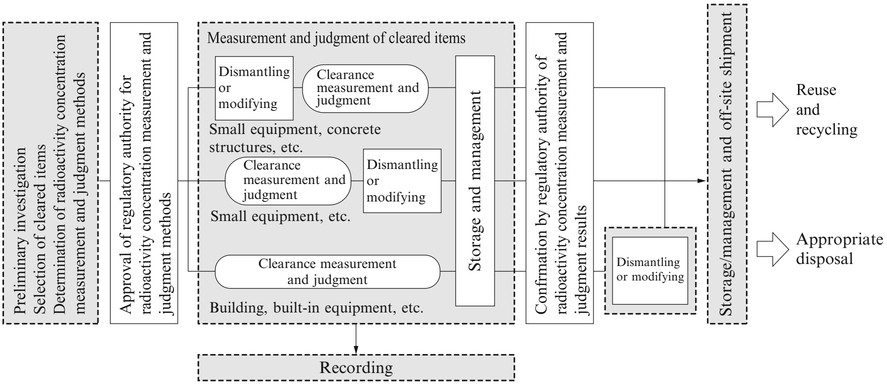


Fig. 4.5 Flow of clearance level verification (example associated with reactor decommissioning)

At the preliminary investigation stage, the state of contamination and various quantities such as the occurrence or nonoccurrence of activated contamination, secondary contamination, and radioactivity levels of cleared items are investigated, and information needed to determine the scope of clearance and conduct measurement and judgment correctly is collected.

At the stage of selection of cleared items, cleared items are selected on the basis of the results of the preliminary investigation, and the cleared items are classified according to the place of occurrence, material, type of contamination and dismantling plans. If necessary, radioactive wastes are separated from cleared items by means of decontamination or scraping.

At the third stage, determination of radioactivity concentration measurement and judgment methods—such as methods for selecting radiation measuring instruments, determining measuring conditions, selecting measuring points and evaluating radioactivity concentration—are determined, and the approval of the regulatory authority is obtained for the decisions thus made.

At the stage of measurement and judgment of cleared items, a dismantling plan appropriate for the characteristics of the cleared items is selected, and, according to that plan, radioactivity concentration is measured by the radioactivity concentration measurement and judgment methods approved by the regulatory authority to determine whether the measured values do not exceed the clearance levels. The records used for the radioactivity concentration measurement and judgment are kept. Furthermore, the regulatory authority checks whether measurement of cleared items and radioactivity concentration judgment have been properly conducted by the approved methods.

At the fifth stage, until the cleared items are being taken out of the site, they are stored and managed properly so as to prevent the intrusion of foreign matter and contaminants.

Thus, the involvement of the regulatory authority in clearance level verification occurs in two stages. At the first stage, examination is made to determine whether details—such as the type of cleared item, radioactivity concentration measurement methods, radiation measuring instruments, measuring conditions, and matters related to management and quality assurance—are appropriate. At the second stage, the records for all cleared items are examined and checked against the items, and sampling inspection is also conducted to check whether the clearance measurement and judgment by the nuclear operator have been carried out by the approved methods. In the sampling inspection, samples are taken from large-quantity cleared items to check on radioactivity concentration and on how the cleared items are stored in containers.

A certification of verification is granted if it has been confirmed by the regulatory authority that the radioactivity concentration of the cleared items is at or below the clearance level and that the clearance measurement and judgment has been carried out by the approved methods.

4.3.2 Clearance Judgment

Following on the description of the concept and methods of clearance level derivation, this section deals with the method by which a nuclear operator can judge whether the cleared items under consideration meet the clearance level criteria. The Standards Committee of the Atomic Energy Society of Japan has specified a number of concrete methods for judging cleared items.

4.3.2.1 Method of Selecting Evaluated Nuclides

As noted at the beginning of this section, the term evaluated nuclide refers to a nuclide whose radioactivity concentration is measured and evaluated for clearance judgment. For reactor facilities, basically, evaluated nuclides are important nuclides. Important nuclides are nuclides having a large *D*/*C* ratio (*D*: estimated radioactivity concentration of the nuclide, *C*: clearance level). The greater this ratio, the more important the nuclide is from the viewpoint of exposure dose evaluation.

As evaluated nuclides for power generation reactor facilities, ten nuclides, namely, ^{3}H, ^{54}Mn, ^{60}Co, ^{90}Sr, ^{134}Cs, ^{137}Cs, ^{152}Eu, ^{154}Eu, ^{239}Pu and ^{241}Am, have been specified in the internal rules [15] of former NISA.

A typical procedure for selecting evaluated nuclides is shown in Fig. 4.6. The procedure has been developed with the aim of identifying all nuclides that

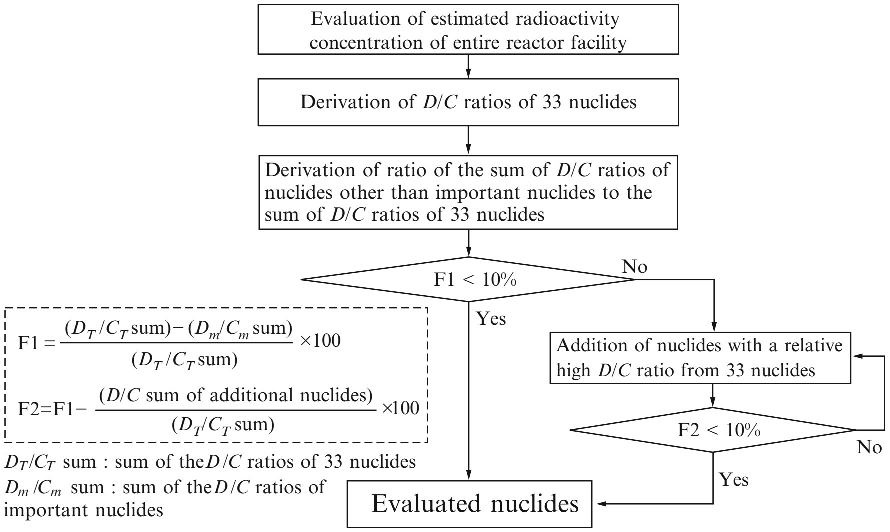


Fig. 4.6 Procedure for selecting evaluated nuclides (*C*: clearance level, *D*: estimated radioactivity concentration of the nuclide)

are important from the viewpoint of exposure dose evaluation among the nuclides that can cause contamination.

The first step is to evaluate the estimated radioactivity concentration D of a total of 33 nuclides that can cause activated contamination or secondary contamination according to the type of reactor, the state of contamination and main materials concerned and then find the D/C ratios of the 33 nuclides by dividing D by the clearance level C of each nuclide. The next step is to derive the sum, D_m/C_m sum, of the D/C ratios of nuclides that are very important in exposure dose evaluation (important nuclides). Then, it is confirmed that the value obtained by subtracting the D_m/C_m sum from the D_T/C_T sum, which is the sum of the D/C ratios of the 33 nuclides, is smaller than 10 % of the D_T/C_T sum (in Fig. 4.6, $\mathrm{F1} < 10\ \%$).

If this condition is satisfied, it is concluded that all important nuclides have been selected, and the selected nuclides are determined as the evaluated nuclides. If the condition is not satisfied, nuclides having a relatively high D/C ratio are chosen from the 33 nuclides and added to the selected nuclides, and the percentage relative to the D_T/C_T sum is derived again to check whether it is smaller than 10 % (in Fig. 4.6, $\mathrm{F2} < 10\ \%$). More nuclides are added, if necessary, until the percentage becomes smaller than 10 %.

4.3.2.2 Radioactivity Concentration Derivation Methods

Methods for deriving the radioactivity concentration of cleared items can be broadly classified into four types: (1) the activation calculation, (2) the radiation measurement method, (3) the radiochemical analysis method, and (4) the theoretical calculation. If the average radioactivity concentration method is used in the radiochemical analysis method, the average radioactivity concentration is derived from the results of the radiochemical analysis performed at the preliminary investigation stage instead of the items used for the clearance measurement conducted at the stage of measurement and judgment of cleared items.

(1) Derivation of radioactivity concentration by the activation calculation

In evaluation made by the activation calculation, the radioactivity concentration of nuclides in the cleared items due to activation is derived by using the neutron fluence rate, activation cross section, elemental composition and neutron irradiation conditions. Further details of the evaluation method are shown in Sect. 3.2.1.

(2) Derivation of radioactivity concentration by the radiation measurement method

In evaluation made by the radiation measurement method, the radioactivity concentration of the cleared item of interest is derived from the counting rate of the radiation measuring instrument for gamma or beta rays and the radioactivity conversion factor.

The radioactivity measurement method includes the key nuclide measurement method, total gamma-ray group measurement method, and the total beta-ray group measurement method. In the key nuclide measurement method, when one nuclide represents a set of count rates, radioactivity is estimated by assuming that all count rates come from that nuclide. The total gamma-ray group measurement method (or the total beta-ray group measurement method) can be applied in cases where there are a number of nuclides emitting gamma rays (or beta rays), and only a total gamma-ray count (or total beta-ray count) can be measured.

In the key nuclide measurement method, if all count rates obtained from a radiation measuring instrument are of gamma or beta rays from a key nuclide, the count rates are converted to radioactivity by using the radioactivity conversion factor for the key nuclide, and radioactivity concentration D_0 [Bq/g] is derived by dividing the converted value by the weight of the cleared item.

$$D_0 = \frac{C_{ts}}{\varepsilon_0} \cdot \frac{K_0}{W_0} \tag{4.5}$$

C_{ts}: count rate [cps],
ε_0: detection efficiency for key nuclide i,
K_0: radioactivity conversion factor for key nuclide [Bq/cps], and
W_0: weight of the cleared item [g].

For a reactor facility, for example, the radioactivity concentration of ^{60}Co is derived by this method. In this case, the radioactivity concentration of other evaluated nuclides is derived by the nuclide composition ratio method or the relative ratio calculation.

If it is thought that there is more than one key nuclide, it is necessary to take into consideration the nuclide composition ratio between the key nuclides. By using the abundance ratio of one of the key nuclides, the nuclide composition ratio and the detection efficiency of each of the evaluated nuclides, the measured count rates are divided proportionally. Then, the radioactivity of each of the evaluated nuclides is derived by using the radioactivity conversion factor, and the radioactivity concentration D_0 of the key nuclide is derived, taking into account the weight of the cleared item as follows.

$$D_0 = C_{ts} \cdot R_0 \cdot \sum \left(\frac{1}{\varepsilon_i} \cdot K_i \cdot \frac{1}{R_i} \right) \cdot \frac{1}{W} \tag{4.6}$$

R_0: abundance ratio of key nuclide,
ε_i: detection efficiency for evaluated nuclide i,
K_i: radioactivity conversion factor for evaluated nuclide i [Bq/cps], and
R_i: nuclide composition ratio between key nuclide and evaluated nuclide i.

Nuclides whose radioactivity concentration is derived by this method include ^{60}Co and ^{137}Cs. In this case, the radioactivity concentration of other evaluated nuclides is derived by the nuclide composition ratio method or the relative ratio calculation.

In the total gamma-ray group measurement method or the total beta-ray group measurement method, all gamma rays or beta rays emitted by the cleared item are measured with a radiation measuring instrument. Then, the count rates thus obtained are converted radioactivity assuming the emission of gamma or beta rays from a single key nuclide, and the radioactivity concentration D_0 of the key nuclide is derived, taking into account the weight of the cleared item.

$$D_0 = \frac{C_{ts}}{\varepsilon_0} \cdot K_0 \cdot \frac{K_{\mathrm{Co}}}{W_0} \tag{4.7}$$

C_{ts}: count rate [cps],
K_0: radioactivity conversion factor for key nuclide [Bq/cps],
K_{Co}: correction coefficient $[(K_i/C_i)/(K_0/C_0)]$,
C_i: clearance level for nuclide i [Bq/g], and
C_0: clearance level for key nuclide [Bq/g].

Here, the correction coefficient is derived from the radioactivity conversion factor and clearance level for an evaluated nuclide emitting beta rays or gamma rays according to the ratio between the D/C ratios of the evaluated nuclide and the key nuclide. For example, the radioactivity concentration of ^{60}Co, ^{134}Cs and ^{137}Cs can be derived by this method. In the total beta-ray group measurement method, the radioactivity concentration of evaluated nuclides that do not emit beta rays is derived by the nuclide composition ratio method, the average radioactivity concentration method or the relative ratio calculation. In the total gamma-ray group measurement method, the radioactivity concentration of evaluated nuclides that do not emit gamma rays is derived in a similar manner.

(3) Derivation of radioactivity concentration by the radiochemical analysis method

For low radioactivity concentration of nuclides that do not allow radioactive nuclide identification or radioactivity concentration estimation by means of radiation measurements, the radiochemical analysis method is used. In this analysis method, samples are taken from the cleared item, and elements are chemically separated for analysis. This analysis method makes it possible to determine the type and radioactivity concentration of nuclides contained in the samples.

Many samples are taken from the cleared items, and the average radioactivity concentration of the cleared items being measured is determined by radioactivity concentration of nuclides contained in the samples from the radiochemical analysis. This method is termed the average radioactivity concentration method. In this method, the radioactivity concentration distribution is assumed to be a logarithmic normal distribution, and a geometric mean is derived from the results of the radiochemical analysis performed at the preliminary investigation stage of the clearance process. The value thus obtained is used as the radioactivity concentration of the evaluated nuclide to be used as a criterion for clearance judgment. The evaluated nuclide to which the average radioactivity concentration method is applied is not used for the clearance measurement conducted at the stage of measurement and judgment of cleared items. If a geometric mean is used, it is necessary to take the uncertainty of radioactivity concentration into consideration. It is also possible to use an arithmetic mean, instead of a geometric mean. In that case, the uncertainty of radioactivity concentration does not need to be taken into consideration because the arithmetic mean tends to be higher than the geometric mean. The radioactivity concentration of ^{3}H can be derived by this method.

(4) Derivation of radioactivity concentration by the theoretical calculation

If there is a correlation between the radioactivity concentration of a key nuclide for which clearance measurements can be made using a radiation measuring instrument and the radioactivity concentration of a evaluated nuclide for which measurements using a radiation measuring instrument is difficult, radioactivity concentration is determined by theoretical calculation using the nuclide composition ratio method or the relative ratio calculation.

In the nuclide composition ratio method, the nuclide composition ratio R_i, which is the ratio between the radioactivity concentration of the key nuclide and that of an evaluated nuclide other than the key nuclide, is determined by using the results of the radiochemical analysis conducted at the preliminary investigation stage. The next step is to determine the radioactivity concentration D_i [Bq/g] of the evaluated nuclide by using R_i as described in Eq. (4.8).

$$D_i = \left(D_0/e^{-\lambda_0 t}\right) \cdot R_i \cdot e^{-\lambda_i t} \tag{4.8}$$

D_i: radioactivity concentration of evaluated nuclide i [Bq/g],
D_0: radioactivity concentration of key nuclide determined through radiation measurement [Bq/g],
R_i: nuclide composition ratio between key nuclide and evaluated nuclide i,
t: duration from reactor shutdown to clearance judgment [y],
λ_i: decay constant for evaluated nuclide i [y^{-1}], and
λ_0: decay constant for key nuclide [y^{-1}].

In the case of a PWR, the radioactivity concentration of ^{54}Mn, ^{90}Sr, ^{134}Cs and ^{137}Cs is determined by using the nuclide composition ratio relative to ^{60}Co.

In the relative ratio calculation, if there is a correlation between the generation behavior of a key nuclide and an evaluated nuclide, radioactivity concentration is determined, using the radioactivity concentration of the key nuclide and physical constants expressing the generation behavior, from the ratio to the radioactivity concentration of the key nuclide. In the case of secondary contamination objects, for example, the relative importance of the D/C ratios of ^{152}Eu and ^{154}Eu is small, and these nuclides are not generated in significant amounts. Radioactivity concentration, therefore, can be determined by the relative ratio calculation by regarding ^{60}Co as the key nuclide.

4.3.2.3 Criterion for Judging Whether Clearance Levels Are Met

The criterion for judging whether the clearance levels are met is whether the sum of the ratios of the radioactivity concentration D_i of the evaluated nuclide i contained in the cleared item to the clearance level C_i for the nuclide is equal to or smaller than 1 as shown in Eq. (4.9).

$$\sum_i \frac{D_i}{C_i} \leqq 1 \tag{4.9}$$

D_i: radioactivity concentration of evaluated nuclide i contained in the cleared item [Bq/g], and
C_i: clearance level for evaluated nuclide i [Bq/g].

Clearance judgments are made in evaluation units, but NISA has its internal rules stipulating that as a general rule, cleared items at a nuclear power plant be evaluated in evaluation units of 1 ton.

4.4 Clearance Applications

In Japan clearance applications are currently being made at the Tokai Power Station of Japan Atomic Power Company (JAPC) with respect to metals and at JRR-3 of the Japan Atomic Energy Agency (JAEA) with respect to concrete.

4.4.1 Clearance Activities at the Tokai Power Station

4.4.1.1 Overview of Clearance Activities

On June 2, 2006, JAPC applied for the clearance of about 2,000 tons of scrap metals as part of the objects to be removed in connection with the dismantling of the refueling equipment and fuel handling building area equipment resulting from the decommissioning of the Tokai Power Station (GCR, 166 MWe). On September 8 of the same year, JAPC was granted approval for the radioactivity concentration measurement and judgment methods.

The scrap metals generated as a result of the decommissioning are cut into pieces small enough to allow them to fit in 1.3 m × 1.3 m × 0.9 m measurement containers. After cutting, grid or shot blasting (mechanical decontamination processes) is carried out for decontamination, and surface contamination density is measured in batches of about 100 kg by using tray-type pre-monitoring system to check for no localized contamination.

Then, about 1 ton of the scrap metals as an evaluation unit is put in a measurement container, and the clearance measurement involving the total gamma-ray count measurement is made to check by the basket-type measuring system in the controlled area if the clearance level is met. Scrap metals that have undergone clearance measurement are stored in a locked and access-controlled confirmation-waiting area located in a non-controlled area until their clearance is confirmed by the regulatory authority.

The basket-type measuring system has been designed specifically for clearance measurement. It uses eight plastic scintillators; two each at the top, bottom, and two sides of the measuring system, and the clearance measurement is done by dividing the entire surface of the container into 40 zones. Besides the clearance measurement in terms of the total gamma-ray counts, the total beta-ray count measurement required to move cleared items from a controlled area to a non-controlled area can also be conducted at the same time.

JAPC, the nuclear operator involved, applied to the regulatory authority for the confirmation of the radioactivity concentration measurement and judgment results for about 107 tons of measured cleared items on April 27, 2007, and for about 291 tons of measured cleared items on March 26, 2008. In response to these applications, the now-defunct Japan Nuclear Energy Safety Organization (JNES), under contract with former NISA, made checks to confirm that the clearance process was carried out in accordance with the approved radioactivity concentration measurement and judgment methods. On May 31, 2007, and May 27, 2008, JAPC was granted certifications of verification of the radioactivity concentration measurement and judgment results by METI.

The 107 tons of cleared metals for which the certification of verification was granted on May 31, 2007, were shipped to the cast product manufacture 1 week later on June 6. This was the first shipment made under Japan's clearance system. After that, the cleared metals were melted by the cast product manufacturer and processed into reception room tables, benches, blocks and shielding materials. These products have been recycled by JAPC, Tokyo Electric Power Company (TEPCO), JAEA, and MEXT.

4.4.1.2 Selection of Evaluated Nuclides

In the clearance activities at the Tokai Power Station, the evaluated nuclides were selected as described below.

The radioactivity concentration associated with activated contamination was derived through activation calculation based on neutron fluence rates, elemental composition and irradiation conditions.

Equipment and devices in systems such as liquid waste and off-gas systems contain liquids and carbon dioxide that include radioactive materials. During the operation of such equipment and devices, radioactive materials attach to or are deposited onto the inside surfaces of the equipment and devices in the systems so as to cause secondary contamination. Therefore, there are three sources of secondary contamination: fission products attributable to nuclear fuel, activated structural materials and activated graphite.

The nuclide composition of each source of contamination is derived through activation calculation. Samples are taken from the equipment and devices in the liquid waste and off-gas systems, and radiochemical analyses are conducted for representative nuclides namely, ^{14}C, ^{60}Co, ^{63}Ni and ^{137}Cs, at each source of contamination. The representative nuclide in the fission products attributable to nuclear fuel is ^{137}Cs, the representative nuclides in the activated structural materials are ^{60}Co and ^{63}Ni, and the representative nuclide in the activated graphite is ^{14}C. Then, the contribution ratios of the three sources of contamination in the liquid waste and off-gas systems are determined from the values obtained through radiochemical analysis and the activation calculation results, and, from the contribution ratios thus obtained, the nuclide compositions in each system is derived. Contamination in all liquid waste and off-gas systems is determined using the nuclide compositions thus obtained.

The next step is to take samples from the inside surfaces of the equipment and devices in the zone being studied for clearance, determine the surface contamination density of representative nuclides such as ^{60}Co, find the ratio between the surface contamination density and nuclide composition of representative nuclides from the derived results, and derive the surface contamination density of the system being studied for clearance.

From these results, a total of 11 nuclides have been selected as evaluated nuclides. These consist of important nuclides, namely, ^{3}H, ^{54}Mn, ^{60}Co, ^{90}Sr, ^{134}Cs, ^{137}Cs, ^{152}Eu, ^{154}Eu, ^{239}Pu and ^{241}Am, as well as ^{14}C, which shows a *D*/*C* sum exceeding 10 %.

4.4.1.3 Radioactivity Concentration Derivation Method

Among the evaluated nuclides, the geometric mean for ^{3}H derived by the average radioactivity concentration method by using the values obtained from the radiochemical analysis conducted at the preliminary investigation stage has been approved as a radioactivity concentration value to be used for clearance judgment. For this reason, ^{3}H is outside the scope of clearance measurement to be made at the stage of measurement and judgment of cleared items. The radioactivity concentration of the other evaluated nuclides is determined by conducting clearance measurement using the basket-type measuring system and using the total gamma-ray group measurement method or the nuclide composition ratio method.

The radioactivity concentration of the cleared item is derived as follows from the results of the clearance measurement conducted using the basket-type measuring system.

First, the radioactivity concentrations of ^{60}Co and ^{137}Cs, which are gamma-emitting nuclides, are determined. The total gamma-ray count rate C_{ts} [cps] is determined from all count results obtained from the measurement using the basket-type measuring system. On the assumption that all of these count values come from ^{137}Cs, the radioactivity concentration D_{Cs} [Bq/g] is derived from Eq. (4.10).

$$D_{\mathrm{Cs}} = C_{ts} \cdot \left(\frac{K_0 \cdot S_0}{\varepsilon_0}\right) \cdot \frac{1}{W_0} \tag{4.10}$$

K_0: radioactivity conversion factor for key nuclide [Bq/cps],
S_0: safety coefficient for key nuclide,
ε_0: detection efficiency for key nuclide, and
W_0: weight of cleared metal [g].

The key nuclide is ^{137}Cs. The radioactivity concentration D_{Cs} [Bq/g] thus obtained is termed the "total gamma-ray radioactivity concentration estimated by ^{137}Cs." The safety coefficient is a factor to allow for non-uniform distribution of count rates in the basket (the measurement container). The count rate in the case where a uniformly distributed radiation source in the measurement container is assumed and the count rate in the case where a radiation source is located at the lowest sensitivity location are determined by using a continuous energy particle transport Monte Carlo code, and the ratio obtained is used as the safety coefficient.

Similarly, the "total gamma-ray radioactivity concentration estimated by ^{60}Co" [Bq/g] is determined by replacing the radioactivity conversion factor for ^{137}Cs with the radioactivity conversion factor for ^{60}Co, the safety coefficient for ^{137}Cs with the safety coefficient for ^{60}Co, and the detection efficiency for ^{137}Cs with the detection efficiency for ^{60}Co, respectively, in Eq. (4.10).

Hence, the radioactivity concentration D_j [Bq/g] of the gamma-emitting evaluated nuclides, namely, ^{54}Mn, ^{60}Co, ^{134}Cs, ^{137}Cs, ^{152}Eu and ^{154}Eu, is determined, by proportionally dividing the total gamma-ray radioactivity concentration estimated by ^{137}Cs according to the relative ratios for these nuclides, as follows:

$$D_j = D_{\mathrm{Cs}} \cdot \frac{R_j}{\sum R_j} \tag{4.11}$$

where R_j is rate of nuclide composition of the gamma-emitting nuclide j, which should have been determined when selecting the evaluated nuclides. The ratio $R_j/\Sigma R_j$ in Eq. (4.11) is called the relative ratio.

The reason why the total gamma-ray radioactivity concentration estimated by ^{137}Cs is used is as follows. Among the gamma-emitting nuclides, ^{137}Cs is the nuclide for which the ratio of the radioactivity conversion factor to the clearance level that takes into account the detection efficiency of the basket-type measuring system turned out to be the highest. The sum of the values obtained by dividing the radioactivity concentration of each of the gamma-emitting nuclides mentioned above by the clearance level, therefore, can be estimated to be higher than in cases where the radioactivity concentrations of other gamma-emitting nuclides are used.

The reason why the total gamma-ray radioactivity concentration estimated by ^{60}Co is used is that ^{60}Co is a gamma-emitting nuclide that provides the correlation needed for deriving the radioactivity concentration of ^{14}C, which does not emit gamma rays; radioactivity concentrations of ^{14}C and ^{60}Co are known to be correlated from the results of the radiochemical analysis conducted on samples taken from the liquid waste and off-gas systems to select evaluated nuclides.

The radioactivity concentration D_{C} [Bq/g] of ^{14}C is derived from the total gamma-ray radioactivity concentration estimated by ^{60}Co and the nuclide composition ratio between ^{14}C and ^{60}Co as Eq. (4.12).

$$D_{\mathrm{C}} = \frac{D_{\mathrm{Co}}}{\exp(-\lambda_{\mathrm{Co}} t)} \cdot R_c \cdot \exp(-\lambda_{\mathrm{c}} t) \tag{4.12}$$

D_{Co}: total gamma-ray radioactivity concentration estimated by ^{60}Co [Bq/g],
R_c: nuclide composition ratio between ^{14}C and ^{60}Co,
t: duration from reactor shutdown to clearance judgment [y],
λ_{c}: decay constant for ^{14}C [y^{-1}], and
λ_{Co}: decay constant for ^{60}Co [y^{-1}].

The radioactivity concentration D_{Sr} of ^{90}Sr, which has a correlation with ^{137}Cs, and the radioactivity concentration D_α of all alpha-emitting nuclides can be derived by replacing D_{Co} in Eq. (4.12) with the total gamma-ray radioactivity concentration estimated by ^{137}Cs, λ_{Co} with the decay constant λ_{Cs} for ^{137}Cs, λ_{c} with the decay constant λ_{Sr} for ^{90}Sr and the decay constant λ_α for ^{239}Pu, R_c with the nuclide composition ratio R_{Sr} between ^{90}Sr and ^{137}Cs, and the nuclide composition ratio R_α between all alpha-emitting nuclides and ^{137}Cs, respectively. The radioactivity concentrations of ^{239}Pu and ^{241}Am are derived by replacing D_{Cs} in Eq. (4.11) with D_α, and using the relative ratios for ^{239}Pu and ^{241}Am, respectively.

Table 4.2 summarizes the radioactivity concentration determination methods for the evaluated nuclides described above. Whether the cleared item under consideration meets the clearance requirements is checked by determining the radioactivity concentration of 10 nuclides from the clearance measurement results by the methods described above, and deriving the D/C ratios for the evaluated nuclides in each of the measurement containers to satisfy the evaluation unit using the average radioactivity concentration of ^{3}H to determine if the sum of the values thus derived is smaller than 1. Uniformity of the radioactivity concentration is checked in batches of a specified size by radiation management record, by surface contamination density measurement using the tray-type pre-monitoring system, or by 40-segment container measurement using the basket-type measuring system.

Table 4.2 Radioactivity concentration determination methods

Nuclide	Evaluation method	Key nuclide	Nuclide composition ratio[a]	Relative ratio[a]	Radioactivity concentration [Bq/g]
^{3}H	Average radioactivity concentration method	–	–	–	9.1×10^{-1}
^{14}C	Nuclide composition ratio method	^{60}Co	7.3×10^{-2}	–	Total gamma-ray radioactivity concentration estimated by ^{60}Co[b] × nuclide composition ratio
^{54}Mn	Total gamma-ray group measurement method	–	–	4.3×10^{-2}	Total gamma-ray radioactivity concentration estimated by ^{137}Cs[c] × relative ratio
^{60}Co	Total gamma-ray group measurement method	–	–	9.1×10^{-1}	Total gamma-ray radioactivity concentration estimated by ^{137}Cs[c] × relative ratio
^{90}Sr	Nuclide composition ratio method	^{137}Cs	1.5×10^{-1}	–	Total gamma-ray radioactivity concentration estimated by ^{137}Cs[c] × nuclide composition ratio
^{134}Cs	Total gamma-ray group measurement method	–	–	8.7×10^{-3}	Total gamma-ray radioactivity concentration estimated by ^{137}Cs[c] × relative ratio
^{137}Cs	Total gamma-ray group measurement method	–	–	3.5×10^{-2}	Total gamma-ray radioactivity concentration estimated by ^{137}Cs[c] × relative ratio
^{152}Eu	Total gamma-ray group measurement method	–	–	2.1×10^{-4}	Total gamma-ray radioactivity concentration estimated by ^{137}Cs[c] × relative ratio
^{154}Eu	Total gamma-ray group measurement method	–	–	1.0×10^{-3}	Total gamma-ray radioactivity concentration estimated by ^{137}Cs[c] × relative ratio
^{239}Pu	Nuclide composition ratio method	^{137}Cs	5.0×10^{-3}[d]	8.2×10^{-1}	Total gamma-ray radioactivity concentration estimated by ^{137}Cs[c] × nuclide composition ratio × relative ratio
^{241}Am	Nuclide composition ratio method			1.8×10^{-1}	Total gamma-ray radioactivity concentration estimated by ^{137}Cs[c] × nuclide composition ratio × relative ratio

Notes

[a] The nuclide composition ratio and the relative ratio are based on those values during reactor shutdown

[b] The total gamma-ray radioactivity concentration estimated by ^{60}Co is the radioactivity concentration derived as the total count rate of gamma rays by using the radioactivity concentration conversion factor for ^{60}Co

[c] The total gamma-ray radioactivity concentration estimated by ^{137}Cs is the radioactivity concentration derived as the total count rate of gamma rays by using a conservative radioactivity concentration conversion factor for ^{137}Cs

[d] The nuclide composition ratio is the ratio in radioactivity concentration of all alpha nuclides (^{239}Pu + ^{241}Am) to the radioactivity concentration of ^{137}Cs

4.4.2 Clearance Activities at JRR-3

4.4.2.1 Overview of Clearance Activities

The modification of JRR-3 carried out by JAEA from 1985 to 1990 generated about 4,000 tons of concrete rubble. An application for the clearance of the concrete rubble was filed in November 2007, followed by an amended application in May 2008, and approval for the clearance was granted on July 25, 2008. The concrete rubble had been stored in twelve 5 m × 10 m × 5 m vaults at the No. 2 Waste Storage Facility in the North Area of the Nuclear Science Research Institute.

Since the concrete rubble included part of the reinforcing bars outside the scope of clearance and combustible wastes that had found their way into the concrete rubble during the modification work, it was necessary, as the first step, to remove them. Reinforcing bars were removed from concrete fragments by using magnetic separators. Concrete blocks were crushed using crushers, and then reinforcing bars were removed with magnetic separators. Concrete fragments from which reinforcing bars had been removed were stored temporally in collecting pallets 25 cm tall and 60 cm in diameter in batches of about 100 kg. Portable Ge semiconductor detectors were then used to inspect each collecting pallet to check if radioactivity concentration was uniformly distributed. The removed reinforcing bars had to be treated as radioactive wastes because there was no recycling plan for them and because they were not covered by the clearance scheme.

The next step was to take powder samples for gamma-ray measurement and fragment samples for ^{3}H measurement from the collecting pallets. Then, concrete fragments from 10 collecting pallets were put into a one-cubic-meter flexible container (evaluation unit) so that each flexible container weighed less than 1 ton.

The grading of powder samples from 10 collecting pallets was adjusted to prepare samples for gamma-ray measurement of each evaluation unit, and measurement was made using a Ge semiconductor detector. Fragment samples were used for measurement by the water immersion method, and the water collected from each evaluation unit was used for ^{3}H concentration measurement with a liquid scintillation counter. The ^{3}H concentration data thus obtained were used to check on the uniformity of radioactivity concentration in a vault.

By using the clearance measurement results, the *D*/*C* ratio of each evaluated nuclide was determined for each of the flexible containers storing 1 ton (evaluation unit) of concrete fragments, and judgment was made as to whether the sum of the ratios met the clearance requirements. All flexible containers satisfying the clearance requirements were stored in temporary storage tents erected in the No. 2 Waste Storage Facility until confirmation was conducted by the regulatory authority. An application for the confirmation of radioactivity concentration measurement and judgment results was filed on January 12, 2010, for about 377 tons of cleared items among the cleared items that had undergone the clearance measurement, and it was confirmed that the clearance activities were carried out in accordance with the radioactivity concentration measurement and judgment methods approved by MEXT. On May 14, 2010, a certification of verification of the radioactivity concentration measurement and judgment results was granted by MEXT. The cleared items, which were then moved to the stock area in the Nuclear Science Research Institute, are expected to be recycled as parking lot base course materials to be used in the Nuclear Science Research Institute. Thus, following the shipment of the cleared items from JAPC, shipment from JAEA was also begun.

4.4.2.2 Selection of Evaluated Nuclides

In the clearance activities for JRR-3, evaluated nuclides were selected as follows.

To select evaluated nuclides, radioactivity concentration for activated contamination and secondary contamination was derived for the 33 nuclides specified in the MEXT ordinance, and nuclides that are important in exposure dose evaluation were selected. The MEXT ordinance, rather than the METI ordinance, is applicable to JRR-3 because it is not a commercial reactor.

As the first step, the radioactivity concentrations of the 33 nuclides in the concrete structures due to activated contamination were derived by using a burn-up calculation code. This required data on the neutron fluence rate, activation cross section, operating time and the elemental composition of the reactor structural materials. The neutron fluence rate was derived using a three-dimensional neutron transport code on Sn method, and neutron group constants necessary for the calculation were determined by a one-dimensional neutron transport code on Sn method.

The source of contamination contributing to secondary contamination are corrosion products leached into heavy water from corroded metals of the reactor tank and reactor cooling system piping, fission products and neutron capture products leached into heavy water from uranium fuel, and ^{3}H produced from heavy water by neutron irradiation. The radioactivity concentration in corrosion products was derived, taking into account the area of contact with heavy water used as moderator and coolant and the corrosion rate, for the aluminum and stainless steel used as reactor tank and piping materials by using the burn-up calculation code. The radioactivity concentration in fission products and neutron capture products was derived with respect to the fission of the uranium fuel leached into heavy water by using the burn-up calculation code.

For the 33 nuclides, the $(D/C)/(\Sigma D/C)$ ratio, which shows the degree of contribution to the exposure dose, was derived from the radioactivity concentration D of each nuclide and clearance level C. According to the derived results, the dominant contributors to activated contamination were ^{152}Eu (51.4 %) and ^{60}Co (41.8 %), while the dominant contributors to corrosion products, fission products, and neutron capture products, which are all produced as a result of secondary contamination, were ^{60}Co (99.5 %) and ^{137}Cs (89.1 %), respectively. On the basis of these evaluation results, four nuclides, namely, ^{3}H, ^{60}Co, ^{137}Cs and ^{152}Eu, were selected as evaluated nuclides which are mixed sources of contamination involving both activated contamination and secondary contamination. Since JRR-3 is a heavy water reactor and ^{3}H is the main source of contamination, ^{3}H was added as one of the evaluated nuclides.

4.4.2.3 Radioactivity Concentration Measuring Method

Radioactivity concentration measurement in the clearance activities for JRR-3 was conducted as follows.

About 50 g of powder sample for gamma-ray measurement and about 50 g of fragments for ^{3}H measurement were taken from on each collecting pallet (100 kg). The grading of powder samples was adjusted so that their particles were not larger than 2 mm. Thus, about 500 g of powder sample for gamma-ray measurement was taken from each 1 ton (evaluation unit) flexible container, and the radioactivity concentrations of ^{60}Co, ^{137}Cs and ^{152}Eu, selected from the evaluated nuclides, were measured with a Ge semiconductor detector. In the measurement conducted on fragmented samples, immersion water collected in the water immersion method by using fragment samples. 5 mL samples for ^{3}H measurement were made from the immersion waters. ^{3}H concentration in 5 mL samples was measured with a liquid scintillation counter.

By using the clearance measurement results, the D/C ratio of each of the evaluated nuclides was derived for each 1 ton (evaluation unit) flexible container to judge whether the sum of the ratios derived met the clearance requirements.

The uniformity of radioactivity concentration distribution of ^{60}Co was verified by measuring the radioactivity concentration of ^{60}Co for each collecting pallet containing about 100 kg of concrete fragments with a portable Ge semiconductor detector and checking if the measured value did not exceed 0.1 Bq/g. The uniformity of radioactivity concentration distribution of ^{137}Cs was judged to be uniformly distributed by checking to confirm that radioactivity concentration of ^{60}Co was uniformly distributed, because it was found at the preliminary investigation stage that radioactivity concentration of ^{137}Cs was lower than radioactivity concentration of ^{60}Co. The uniformity of radioactivity concentration distribution of ^{3}H was verified by checking if radioactivity concentration of ^{3}H in each vault was lower than 100 Bq/g (clearance level) by using the results of the measurement on the samples for ^{3}H measurement prepared by collecting the immersion water of the fragment samples taken from the collecting pallets. Uniformity verification for ^{152}Eu was not made because contamination by ^{152}Eu is due to activated contamination and radioactivity concentration distribution does not vary significantly.

4.5 Clearance Level Approaches of International Organizations

4.5.1 International Atomic Energy Agency

In Safety Series SS-89 [12], IAEA indicated standards for risks to individuals and the optimization of radiation protection as basic standards for exemption. According to the standards, the criteria for exemption are that the radiation dose is of the order of a few tens of μSv/y in order to keep radiological risks to individuals sufficiently low. IAEA thinks that the optimization of radiation protection aiming to keep the radiation dose as low as reasonably achievable, economic and social factors being taken into account can be achieved if the collective dose commitment is lower than 1 man-Sv/y.

In 1996, IAEA proposed a set of unconditional clearance levels in the technical document TECDOC-855 [2] and indicated for the first time the concept of unconditional clearance levels and derivation methods for them for application to relatively large quantities of wastes and recyclable solid materials generated at reactor facilities.

Concerning clearance levels for gaseous, liquid and solid materials at hospitals, research institutions and industrial facilities, IAEA released the technical document TECDOC-1000 [16] in 1998. In that document, clearance levels for 33 nuclides in use in the areas of medicine and research were derived. According to the document, for solid materials, the exemption level may be used as a clearance level if only moderate quantities (or the order of 1 ton or less per year) are involved. If larger quantities are involved, 1/10 of the exemption level may be used as the clearance level.

At its general conference in September, 2000, IAEA adopted a resolution concerning the determination of criteria for radioactivity concentrations of nuclides in goods for smooth international trade of goods from areas contaminated by a nuclear accident. In August, 2004, the IAEA Safety Guide RS-G-1.7 [7] was completed.

The IAEA Safety Guide RS-G-1.7 derives exemption levels for nuclides that may be exempted in connection with large quantities of solid materials other than foodstuffs and drinking water on the basis of the concept of exclusion with respect to naturally occurring nuclides and the concept of exemption with respect to artificial nuclides. The IAEA Safety Guide RS-G-1.7 states that these exemption levels may be used as clearance levels for similar materials. In determining reference dose values, the IAEA Safety Guide RS-G-1.7 also states that the radiological basis for determining exemption levels for artificial nuclides is that individual effective dose should be of the order of 10 μSv/y or less.

In order to derive exemption levels for artificial nuclides from reference dose values, evaluation pathways associated with recycling and reuse and burial disposal have been studied to investigate the influence of radiation from artificial nuclides. Scenario parameters in such evaluations have been defined conservatively in view of the fact that the natural environment and people's life styles vary widely among different parts of the world.

The exemption levels for naturally occurring nuclides have been determined taking into consideration the global distribution of radioactivity concentration in environmental substances.

The derived exemption levels are processed on a logarithmic basis and shown as representative values expressed in the form 1×10^n. The exemption levels are sufficiently lower than the natural radiation levels to which people are normally exposed. When each country takes regulatory measures, therefore, radioactivity concentrations of the order of up to 10 times the exemption levels may be used as exemption levels or clearance levels at the discretion of the regulatory authority.

If the radioactivity concentration of an object is higher than the exemption level, the following regulatory measures can be taken:

1. If radioactivity concentration exceeds the exemption level by several times, the regulatory authority may decide not to apply regulatory requirements depending on the framework of the domestic regulatory control. In such a case, a decision is made on a case-by-case basis, but the regulatory authority may stipulate in advance that such levels of radioactivity concentration do not need to be regulated
2. If the regulatory authority has decided to take regulatory measures, the degree of stringency of regulatory measures associated with notification and licensing should be commensurate with the level of risk involved.

This approach is called the graded approach.

4.5.2 European Commission

In 1988, with the aim of establishing common standards in the European Union (EU) region, EC made recommendations on clearance levels for recycling for materials containing alpha-emitting nuclides and materials containing beta and gamma-emitting nuclides [17]. In EC, a group of experts formed in 1990 under Article 31 of the Euratom Treaty defined the concepts of action and clearance in 1996 [18]. The group concluded that if the radioactivity concentration of nuclides contained in materials generated from regulated practices is below the clearance levels, the regulatory authorities may exempt the materials from the regulatory requirements for recycling, reuse or disposal. The group also recommended that the regulatory authority of each country base its decisions on the EC guidance when deciding on clearance levels.

As reference dose values used for clearance level derivation, EC uses an individual effective dose of 10 μSv/y, an equivalent dose for the skin of 50 mSv/y, and a collective dose commitment of 1 man-Sv/y.

Because goods to be recycled are expected to be distributed in the EU region as a result of exemption, a systematic series of exemption studies were conducted and the guidances shown in Fig. 4.7 were recommended.

The recommended criteria for the recycling of metals, namely, iron, iron alloys, aluminum, aluminum alloys, copper, copper alloys, as well as the reuse of equipment were released in 1998 as Guidance RP89 [19].

The recommended criteria for the disposal of concrete and the reuse of buildings were released as Guidance RP113 [20] in 2000. Other recommendations include Guidance RP122 Part I (general clearance levels applicable to all solid materials) and Guidance RP122 Part II (clearance levels applicable to naturally occurring radioactive materials).

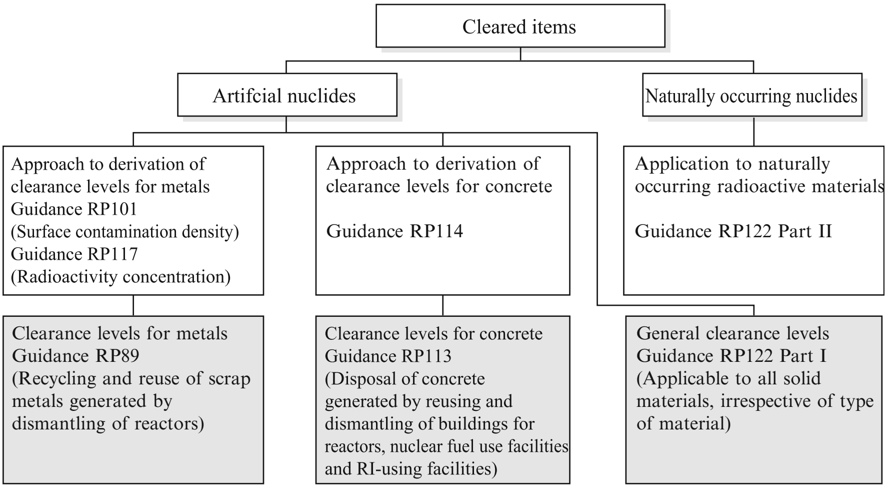


Fig. 4.7 European Commission's system of recommendations on clearance

4.6 Clearance for Non-reactor Facilities

4.6.1 Clearance for Uranium-Handling Facilities

International discussions on clearance for uranium have been underway on the basis of the fact that ^{234}U, ^{235}U and ^{238}U are naturally occurring nuclides. The IAEA Safety Guide RS-G-1.7 specifies clearance levels for naturally occurring nuclides separately from those for artificial nuclides. Table 4.3 shows the clearance levels of radioactivity concentration of uranium nuclides specified in the IAEA Safety Guide RS-G-1.7. As shown, the clearance levels for ^{232}U and ^{236}U, which are artificial nuclides, are 0.1 Bq/g and 10 Bq/g, respectively, and the clearance level for naturally occurring nuclides other than ^{40}K is 1 Bq/g.

In view of the concepts indicated in the IAEA Safety Guide RS-G-1.7, the former NSC of Japan has derived clearance levels for metals used at uranium-handling facilities [21]. If it is assumed that the clearance level for wastes generated by the operation and dismantling of uranium-handling facilities is 1 Bq/g, which is the radioactivity concentration specified in the IAEA Safety Guide RS-G-1.7, metals will account for about 90 % of all cleared items. Cleared items, therefore, have been limited to metals. Nuclear operators generating the greatest quantities of cleared items will generate about 20,000 tons in 10 years. The average quantity of cleared items generated, therefore, is assumed to be 2,000 tons per year. Unlike the cleared items associated with reactor facilities and nuclear fuel use facilities, cleared items here do not include "non-radioactive wastes."

There are five evaluated nuclides, namely, ^{232}U, ^{234}U, ^{235}U, ^{236}U and ^{238}U. These are the results of evaluation based on derived values for uranium conversion and enrichment facilities, measured values obtained from various facilities,

Table 4.3 Determined clearance levels for uranium

Nuclide	NSC of Japan	IAEA RS-G-1.7		METI[a] Uranium processing facilities	MEXT[b] Uranium-using facilities
	Clearance level	Artificial nuclide	Naturally occurring nuclide[c]	Clearance level	Clearance level
^{232}U	0.1	0.1	–	0.1	0.1
^{234}U	1	–	1	1	1
^{235}U	1	–	1	1	1
^{236}U	1	10	–	10	10
^{238}U	1	–	1	1	1

[a] Uranium Clearance Working Group, Radioactive Waste Safety Subcommittee, Nuclear and Industrial Safety Subcommittee, Advisory Committee for Natural Resources and Energy, METI

[b] Technology Working Group, Research Reactor Safety Regulation Committee, MEXT

[c] 1 Bq/g for naturally occurring nuclides other than ^{40}K

enrichment factors achieved at fuel fabrication facilities, uranium specifications at the time of receipt and the standards to be relied on. As in the cases of reactor facilities and nuclear fuel use facilities, the reference dose values used for clearance level derivation are an individual effective dose of 10 μSv/y and an equivalent dose for the skin of 50 mSv/y.

For the purpose of evaluation, evaluation pathways related to the recycling of metals have been extracted from the 92 evaluation pathways for reactor facilities and nuclear fuel use facilities. In view of the fact that uranium, when melted, tends to be transferred to slag, the recycling of slag as parking lot base course materials has been added to the evaluation pathways. Also, because only external exposure had been taken into consideration in evaluating external exposure during parking lot construction, dust inhalation, ingestion intake and skin exposure have been added. Thus, evaluation has been made for a total of 41 evaluation pathways.

The exposure dose evaluation models and parameter values are identical to those used in connection with reactor facilities and nuclear fuel use facilities. When periods of the order of several tens of thousand years are considered for evaluation, exposure doses are expected to increase because of the generation of progeny nuclides of ^{234}U, ^{235}U and ^{238}U. In view of the fact, however, that the service life of metal products is shorter than 100 years, the evaluation period for the maximum exposure dose has been set at 100 years.

In response to the results of the NSC of Japan study on clearance levels for uranium, the Technology Working Group [22], Research Reactor Safety Regulation Committee, MEXT deliberated on clearance for uranium-using facilities, and the Uranium Clearance Working Group [23], Radioactive Waste Safety Subcommittee, Nuclear and Industrial Safety Subcommittee, Advisory Committee for Natural Resources and Energy, METI deliberated on clearance for uranium processing facilities. For the purposes of the deliberations, it was assumed that even if there is similarity, nuclides regulated under the Reactor Regulation Act should be considered separately from naturally occurring nuclides and should be treated in the same manner as artificial nuclides.

Because neither the IAEA Safety Guide RS-G-1.7 nor Safety Reports Series No. 44 [24], a report describing the basis for radioactivity concentration derivation, deals with the derivation of radioactivity concentrations corresponding to reference dose values based on the exposure dose derivation of ^{234}U, ^{235}U and ^{238}U, METI derived clearance levels for cases using the evaluation pathways and evaluation models used in the evaluation described in Safety Reports Series No. 44.

As shown in Table 4.3, the clearance levels adopted by both METI and MEXT are 0.1 Bq/g for ^{232}U, 10 Bq/g for ^{236}U and 1 Bq/g for ^{234}U, ^{235}U and ^{238}U. As for the criteria for judging whether clearance levels are met, it is reasonable to check whether the sum of the values obtained by dividing the average radioactivity concentration of each of the evaluated nuclides by the clearance level is not greater than 1. The reason for this is that unlike naturally occurring nuclides that are in radioactive equilibrium, ^{234}U, ^{235}U and ^{238}U used at facilities have been mostly rid of progeny nuclides in the refining process, and therefore it is appropriate to handle them in the same manner as ^{232}U and ^{236}U, which are of artificial origin.

4.6.2 Clearance for Radioisotope-Using Facilities and Radiation Generators

The clearance system for radioactive wastes under the Reactor Regulation Act has been described in the preceding sections. A clearance system is also introduced into the Law Concerning Prevention from Radiation Hazards due to Radioisotopes, etc. In Japan, there are many radioisotope (RI)-using facilities under the management of operators that use RIs (RI operators), such as medical institutions, research institutions, educational institutions and private-sector enterprises, in many parts of the country. Materials contaminated with RIs (RI-contaminated materials) are stored in these facilities. There are also medical linear accelerators, small cyclotrons for producing nuclides for positron emission tomography (PET) and synchrotrons in use at medical institutions, research institutions, educational institutions and private-sector enterprises. The dismantling of these devices generates activated materials.

Clearance levels are specified, therefore, for RI-contaminated materials and materials contaminated with radiation generated by radiation generators. Unlike cleared items associated with reactor facilities or nuclear fuel use facilities, cleared items associated with RI-using facilities or radiation generators include incineration ash in addition to concrete and metals. Consequently, incineration has been added to burial disposal, recycling and reuse as evaluation pathways for cleared items, and evaluation pathways leading to it have also been defined. Reference dose values are 10 μSv/y as an effective dose to the general public and workers and 50 mSv/y as an equivalent dose for the skin.

The exposure pathway identification process is somewhat special. It is assumed that materials generated at RI-using facilities in Japan and then collected in a year, materials already kept in storage by the Japan Radioisotope Association and materials generated by JAEA are cleared collectively and then buried at one waste disposal facility or transported to one recycling facility for the purpose of recycling. It is also assumed that materials generated at RI-using facilities are cleared individually by each RI operator and then the cleared materials are buried at one waste disposal facility or transported to one recycling facility for the purpose of recycling.

Thus, in view of the current practices and actual quantities of RI-contaminated materials, diverse evaluation pathways applicable to different quantities of cleared items have been defined, including individual clearance in which materials are cleared on an operator-by-operator basis and collective clearance in which materials are collected and transported by waste collectors and cleared collectively.

Exercises

1. Derive the radioactivity concentration of ^{60}Co corresponding to a reference dose value when cleared iron is to be melted, fabricated and recycled as frying pans. Assume a reference dose value of 10 μSv/y. Consider ingestion intake for exposure dose evaluation, and derive exposure doses to adults of the general public. Assume the following: the weight ratio of the cleared metals in the metals to be recycled, 0.1; the density of iron, 7.86×10^6 g/m^3; the ion corrosion rate, 1.3×10^{-4} m/y; the surface area of a frying pan, 7.07×10^{-2} m^2; annual cooking time involving the use of the frying pan, 180 h; dose conversion factor for ingestion intake of ^{60}Co by an adult, 3.49×10^{-9} Sv/Bq; the transfer factor of ^{60}Co to the frying pan in the melting process, 1; the duration from clearance to recycling, 1 year; the exposure time during the exposure scenario, 1 year; and the dilution factor for the recycled metals, 1.
2. Derive the radioactivity concentration of ^{60}Co corresponding to a reference dose value when cleared concrete is to be recycled as coarse aggregate for parking lot construction. Assume a reference dose value of 10 μSv/y. Consider external exposure for exposure dose evaluation, and evaluate exposure doses to adults of the general public. Assume the following: the weight ratio of the cleared concrete in the concrete to be recycled, 0.1; the ratio between the bitumen and the recycled coarse aggregate, 0.25; the time of use of the parking lot in a year, 1,000 h; dose conversion factor for external exposure, 1.9×10^{-3} (Sv/y)/(Bq/g); the duration from clearance to recycling, 0 years; and exposure time during the exposure scenario, 1 year.

References

1. Radioactive Waste Safety Subcommittee, Nuclear and Industrial Safety Subcommittee, Advisory Committee for Natural Resources and Energy, Ministry of Economy, Trade and Industry, *Report on Handling of Non-radioactive Wastes at Nuclear Facilities* (2007) (in Japanese)
2. International Atomic Energy Agency, *Clearance Levels for Radionuclides in Solid Materials: Application of Exemption Principles*, Interim Report for Comment, IAEA TECDOC-855 (1996)

3. Nuclear Safety Commission of Japan, *Clearance Levels for Major Reactor Facilities* (1999) (in Japanese)
4. Nuclear Safety Commission of Japan, *Clearance Levels for Heavy Water Reactors, Fast Reactors, etc.* (2001) (in Japanese)
5. Nuclear Safety Commission of Japan, *Clearance Levels for Nuclear Fuel Use Facilities* (Facilities Dealing With Irradiated Fuels and Materials) (2003) (in Japanese)
6. International Atomic Energy Agency, *International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources*, IAEA Safety Series No. 115 (1996)
7. International Atomic Energy Agency, *Application of the Concepts of Exclusion, Exemption and Clearance*, IAEA Safety Guide No. RS-G-1.7 (2004)
8. Nuclear Safety Commission of Japan, *The Concept of Clearance Level Verification for Reactor Facilities* (2001) (in Japanese)
9. Nuclear Safety Commission of Japan, *Radioactivity Concentration of Items That Do Not Require Handling as Radioactive Wastes Generated by the Dismantling of Reactor Facilities and Nuclear Fuel Use Facilities* (2004) (in Japanese)
10. Radioactive Waste Safety Subcommittee, Nuclear and Industrial Safety Subcommittee, Advisory Committee for Natural Resources and Energy, Ministry of Economy, Trade and Industry, *Establishment of the Clearance System for Nuclear Facilities* (2004) (in Japanese)
11. Reactor Safety Regulation Committee, Ministry of Education, Culture, Sports, Science and Technology, *The Concept of Safety Regulation for Research Reactor Facilities* (2005) (in Japanese)
12. International Atomic Energy Agency and Nuclear Energy Agency of the Organisation for Economic Co-operation and Development, *Principles for the Exemption of Radiation Sources and Practices from Regulatory Control*, IAEA Safety Series No. 89 (1988)
13. International Committee on Radiation Protection, *Protection of the Public in Situations of Prolonged Radiation Exposure*, ICRP Publication 82, Ann. ICRP **29**(1–2), (1999)
14. A.V. Pinner, C.R. Hemming and M.D. Hill, *An Assessment of Radiological Protection Aspects of Shallow Land Burial of Radioactive Wastes*, NRPB-R-161 (1984)
15. Radioactive Waste Regulation Division, Nuclear and Industrial Safety Agency, Ministry of Economy, Trade and Industry, *Approval for Radioactivity Concentration Measurement and Evaluation Methods (Internal Rules)*, NISA-197c-06-1 (2006) (in Japanese)
16. International Atomic Energy Agency, *Clearance of Materials Resulting from the Use of Radionuclides in Medicine, Industry and Research*, IAEA-TECDOC-1000 (1998)
17. European Commission, *Radiation Protection Criteria for the Recycling of Materials from the Dismantling of Nuclear Installations*, RP43, Luxembourg (1988)
18. Commission of the European Communities, *Basic Safety Standards for the Health Protection of the General Public and Workers against the Dangers of Ionizing Radiation*, Council Directive 96/29/EURATOM, Luxembourg (1996)
19. European Commission, *Recommended Radiological Protection Criteria for the Recycling of Metals from the Dismantling of Nuclear Installations*, RP89, Luxembourg (1998)
20. European Commission, *Recommended Radiological Protection Criteria for the Clearance of Buildings and Building Rubble from the Dismantling of Nuclear Installations*, RP113, Luxembourg (2000)
21. Nuclear Safety Commission of Japan, *Clearance Levels for Uranium-handling Facilities* (2009) (in Japanese)
22. Technology Working Group, Research Reactor Safety Regulation Committee, Ministry of Education, Culture, Sports, Science and Technology, *Verification of Clearance Levels for Uranium-handling Facilities* (2010) (in Japanese)
23. Uranium Clearance Working Group, Radioactive Waste Safety Subcommittee, Nuclear and Industrial Safety Subcommittee, Advisory Committee for Natural Resources and Energy, Ministry of Education, Culture, Sports, Science and Technology, *Establishment of the Clearance System for Uranium-handling Facilities* (2010) (in Japanese)
24. International Atomic Energy Agency, *Derivation of Activity Concentration Values for Exclusion, Exemption and Clearance*, IAEA Safety Reports Series No. 44 (2005)

Chapter 5
Radioactive Waste Treatment Technologies

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5.1 Principles of Radioactive Waste Treatment

As discussed in Sect. 1.3.1 "Radioactive waste treatment and disposal processes," the treatment of radioactive wastes includes three steps: pretreatment, treatment and conditioning; these three processes are not performed independently of each other. Instead, appropriate pretreatment, treatment and conditioning methods need to be selected based on the characteristics of the wastes to be treated (e.g., the amount, physical and chemical properties, contained nuclides, and radioactivity level) and also by taking into account both the interconnectedness of the individual steps of the treatment processes and the disposal process subsequent to the treatment processes. These matters are discussed as Principle 8 "Radioactive Waste Generation and Management Interdependencies" of the IAEA's *Principles of Radioactive Waste Management Safety Fundamentals* [1].

Safe and economical management (treatment and disposal) of radioactive waste has been a longstanding issue in promoting the use of radioactive materials as well as the development and use of nuclear energy. Accordingly, efforts and achievements have been made proactively to improve existing technologies and introduce new technologies with emphasis on the following goals [2]: (1) measures for reduction of secondary radioactive waste generation; (2) reduction of radioactivity released to the environment; (3) volume reduction of generated waste; (4) stabilization of waste forms to be disposed of; and (5) reduction of radiation exposure of workers.

The nuclear regulatory system in Japan has been changed significantly after the Fukushima Daiichi Nuclear Power Station accident in March 2011. Descriptions in this chapter have been translated from the book originally published in Japanese before the accident, with minimal update where appropriate.

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The current trend is that generated radioactive wastes are treated for concentration and confined to minimize the release of radioactive materials into the environment. For this reason, instead of merely solidifying generated liquid wastes and other materials, barrier functions are called for so that the safety of treatment is enhanced and the buried disposed waste forms themselves do not easily leach their radionuclides into the groundwater. Accordingly, stabilizing treatment technologies that take into account long-term soundness are already necessary at the stage of waste treatment. In addition, from the perspective of effective use of the limited volume of the burial disposal facility, volume reduction processing has been proactively applied to radioactive wastes.

In the process of treating radioactive wastes, operations are performed such as reduction of generated wastes, removal of radionuclides contained in the wastes, and modification of the waste composition. In principle, these methods are identical to those used for ordinary treatment of industrial waste. As for the characteristics of the radioactive waste treatment, the aim is to minimize the volume of the radioactive secondary wastes generated in the course of the treatment; however, attention should be paid to preventing radiation exposure of workers by such means as confining radionuclides and shielding workers from radiation exposure. This chapter discusses the principles of major treatment methods that depend on the properties of the radioactive wastes to be treated, placing emphasis on these characteristics.

5.2 Gaseous Waste Treatment

Gaseous wastes are treated in the same way as liquid wastes which allows minimization of the amount of radioactivity released into the environment. Therefore, for gaseous waste treatment, the treatment methods, the treatment capacity and the stack height need to be designed by sufficiently taking into account factors such as gasification characteristics of the radionuclides to be used as well as geographical and meteorological conditions around the treatment facility.

The radioactive materials that should be considered as gaseous wastes are: (1) radioactive noble gases such as Kr and Xe; (2) radioactive I (iodine); and (3) particulate matter. Typical methods of treating these kinds of gaseous wastes are filtration (adsorption), attenuation, and dilution. An overview of the typical treatment methods of these radioactive materials is given below.

5.2.1 Radioactive Noble Gases

Radioactive noble gases that are representative of those contained in the off-gas from a nuclear reactor are Kr and Xe. Their major isotopes include: ^{85m}Kr (half-life: 4.4 h), ^{87}Kr (78 min), ^{88}Kr (2.77 h), ^{133}Xe (5.27 day), ^{133m}Xe (2.3 day), ^{135}Xe (9.13 h) and ^{138}Xe (17 min); and many are relatively short lived. These relatively short-lived noble gases are treated by combining the attenuation effect due to their short half-lives and dilution. Because BWRs produce a large amount of gaseous wastes that contain noble gases, the noble gas hold-up system is utilized to treat the gaseous waste generated from them. In this system, noble gases are selectively stored in activated charcoal over a certain time period in order to sufficiently attenuate radioactivity. Subsequently, after confirming that the radioactivity concentration of the noble gases is sufficiently below the regulation limit, they are released into the atmosphere.

Adsorption onto activated charcoal is physisorption, and the adsorption increases as the boiling point of a substance is higher (Xe, −108.1 °C; Kr, −153.4 °C; O_2, −182.9 °C; N_2, −195.8 °C; H_2, −252.8 °C). When off-gas is passed through an activated charcoal bed, most of the constituents of air, e.g., O_2 and N_2, which have low boiling points, pass through the bed without undergoing any changes. On the other hand, Kr and Xe, which have higher boiling points, move through the activated charcoal bed while undergoing reversible adsorption; in the course of this process, radioactivity is attenuated.

If adsorption of the noble gases onto activated charcoal is assumed to be in equilibrium, the relationship among the number of theoretical stages of the noble gas hold-up system, N, hold-up time, t_m [h], the mass of the activated charcoal, W [ton], and the flow rate of the gas to be treated, F [m^3/h], can be expressed as Eq. (5.1): [3]

$$t_m = \frac{N-1}{N} \cdot \frac{K \cdot W}{F} \tag{5.1}$$

where K is known as the dynamic adsorption equilibrium constant [m^3/ton], and its value changes according to the physical properties of the activated charcoal, composition of the gas, pressure, temperature, and some other variables. In particular, the value of K increases with decreasing temperature. At room temperature and pressure, K approximately ranges from 50 to 80 and from 400 to 1,000 for Kr and Xe, respectively. As an example, for a 1,000 MWe-class nuclear reactor, the mass of activated charcoal and flow rates of the gas are 70 tons and 40 m^3/h, respectively, and the hold-up times for Kr and Xe are approximately 40 h and 27 day, respectively.

The number of theoretical stages, N, is in practice approximately 10, which is a large value. Furthermore, the adsorption capacity of activated charcoal is influenced highly by water vapor: the adsorption capacity of activated charcoal with a moisture content of 10 % decreases to approximately half of that of dry activated charcoal. Therefore, a dehumidifier is necessary as auxiliary equipment in the hold-up system. The relationship between the radioactivity concentrations at the inlet and outlet of the noble gas nuclide in the hold-up system, A_i^{in} and A_i^{out}, respectively, and the hold-up time, t_m^i, can be expressed as

$$\frac{A_i^{\text{out}}}{A_i^{\text{in}}} = \exp\left(-\lambda_i t_m^i\right) \tag{5.2}$$

where λ_i is the decay constant of radionuclide i.

5.2.2 Radioactive Iodine

Radioactive iodine that is contained in the off-gas from nuclear facilities takes various chemical forms, which include elemental iodine (I_2), iodic acids (HOI, HOI_3 and HIO_4), and organic iodine compounds (CH_3I, C_2H_5I). Removal of elemental iodine by activated charcoal is relatively easy, and this method is used to remove ^{131}I (8.02 day). As for the adsorption of elemental iodine onto activated charcoal, Eq. (5.3) which assumes that the rate of adsorption is controlled by gas diffusion through a boundary film has been confirmed to agree well with experimental results.

$$\eta = \frac{C_{\text{in}} - C_{\text{out}}}{C_{\text{in}}} = 1 - \exp\left(-K_C \cdot A_S \cdot \frac{L}{U}\right) \tag{5.3}$$

C_{in}, C_{out}: concentration of radioactive iodine at the filter inlet and outlet [Bq/m^3]
η: collection efficiency
K_C: gas film mass transfer coefficient [m/s]
A_S: surface area of the loaded activated charcoal particles [m^2/m^3-activated charcoal]
L: thickness of the activated charcoal bed [m]
U: filtration rate [m/s]

Organic iodine compounds such as methyl iodide (CH_3I) are more volatile than elemental iodine. The iodine filtration method removes radioactive iodine using this volatility property and the isotope exchange reaction. Iodine filters are impregnated with activated charcoal to which small amounts of potassium iodide and iodine were added. Radioactive iodine is exchanged for the non-radioactive iodine (*I) that is impregnated on the filter as expressed by the reaction formula below and can be collected by an iodine filter.

$$\text{KI(activated charcoal)} + \text{CH}_3{*}\text{I} \rightarrow \text{K}{*}\text{I(activated charcoal)} + \text{CH}_3\text{I}$$

The off-gas which is generated at reprocessing facilities and contains ^{129}I (15.7 million years) is scrubbed with an alkaline solution or is put through a solidification using silver. In the alkaline solution scrubbing treatment, the off-gas is scrubbed using a bubbling process in a sodium hydroxide solution, which allows absorption and removal of the iodine in the form of NaI. However, organic iodine is not removed by this treatment. In the solidification using silver, silver is either coated onto or immersed into zeolite, silica gel, aluminum oxide or other substances. The resulting silver-containing materials are filled and heated at approximately 150 °C in columns, through which the off-gas is passed. This procedure solidifies volatile iodides of all forms as stable silver iodide.

5.2.3 *Particulate Matter*

At nuclear facilities, particulates of submicrons to a few microns in size are the targets to be captured in most cases. Among these particulates, coarse particulates are captured by pre-filters while fine particulates are captured by high efficiency particulate air (HEPA) filters.

Pre-filters are also known as coarse particle filters and capture particles of approximately 50 μm in particulate diameter. HEPA filters are guaranteed for their performance to capture dioctyl phosphate particles of 0.3 μm in diameter at an efficiency of 99.97 % or higher at the rated airflow rate. In order to increase the filtration area (up to 50 times the apparent area), the filter paper which is made of fibrous glass is folded in a corrugated manner and is encased in a rigid frame. Fibrous filter materials such as those of HEPA filters capture aerosol particles using the diffusive and inertial nature of particle motions and the screening effect of the filter. The same materials capture coarse particles, relying mainly on the inertial collision of the particles with the filter fibers. As for the particles of a tenth of a micron or smaller in size, they are captured by the fibrous filter material upon colliding with the fibers due to diffusion caused by Brownian motion

The capture efficiency of HEPA filters, E, is calculated using the measured value of the aerosol concentration at the filter inlet, C_{in}, and that at the filter outlet, C_{in}, as in the following equation.

$$E = \frac{C_{\text{in}} - C_{\text{out}}}{C_{\text{in}}} \tag{5.4}$$

Filters used for clean rooms are called ultra low penetration air (ULPA) filters, and their capture efficiency is even higher than that of HEPA filters. Specifically, the regulation stipulates that ULPA filters be able to capture particles of 0.15 μm in diameter at the capture efficiency of 99.9995 % or higher at the rated airflow rate and that the initial pressure drop of the ULPA filters be 245 Pa or lower.

As a reference, the sizes of some of the particles that are encountered in daily life are as follows: 30 μm for pollen, 5–10 μm for mold spores, 0.01–10 μm for cigarette smoke particles, and 1 μm or smaller for flu viruses. Therefore, HEPA filters are able to remove pollen and mold spores, but only some of the cigarette smoke particles. HEPA filters are ineffective at removing flu viruses.

5.3 Liquid Waste Treatment

Treatment of radioactive liquid wastes is a process of separation and concentration of radionuclides. The concentrates that are produced from this process and undergo volume reduction are treated into a stable waste form with a method such as those described in Sect. 5.5, “Solidification treatment.” After the concentration of the

Table 5.1 Liquid waste treatment methods

Treatment method Properties of liquid waste		Coagulation-sedimentation	Filtration	Ion exchange	Evaporation
Amount of ions [μS/cm]	High (a few hundred or higher)	Appropriate	Inappropriate	Inappropriate	Appropriate
	Low (a few tens or less)	Inappropriate	Inappropriate	Appropriate	Inappropriate
Amount of crud [ppm]	High (a few hundred or higher)	Appropriate	Possibly appropriate	Inappropriate	Possibly appropriate
	Low (a few tens or less)	Possibly appropriate	Appropriate	Inappropriate	Inappropriate

diluted liquid is confirmed to be below the regulation limits, the diluted liquid is released into the environment.

Treatment methods of liquid waste can be classified roughly into coagulation-sedimentation (adsorption), ion exchange, filtration, and evaporation [4]. An appropriate treatment method is selected according to the liquid waste to be treated, that is, by taking into account the characteristics of the liquid waste (e.g., the radioactivity level, the amount of dissolved ions and the amount of solid materials), the regulation limits for the decontaminated liquid waste, the conditioning method for the concentrate generated from the treatment, and some other factors.

Table 5.1 shows general comparisons of the above-mentioned treatment methods. Amounts of ions and crud in liquid waste are two major criteria in selecting the treatment method. The amount of ions corresponds to the amount of dissolved materials, that is, concentration of ions in the liquid, and is expressed with the unit of conductivity (μS/cm). The amount of crud corresponds to the amount of insoluble solids and is expressed as its concentration (ppm). Because the coagulation-sedimentation produces precipitates by chemical reactions and separates them from the liquid waste, it is not well-suited for low concentration liquid waste that contains a small amount of ions. The filtration is intended to remove solid constituents from liquid waste. Therefore, regardless of the amount of ions present in the liquid waste, the principle behind this method does not allow its application to liquid containing only dissolved constituents.

On the other hand, the ion exchange is aimed at removing ions, that is, dissolved constituents. Therefore, the principle behind this method does not allow crud removal. Although the ion exchange is designed for ion removal, efficient ion removal cannot be expected from highly concentrated liquid waste, that is, liquid waste with high ion content, because of the limit on the ion exchange capacity. As with the coagulation-sedimentation, the evaporation is applicable to both dissolved and insoluble constituents. Nonetheless, this method is inefficient unless the concentration of the liquid waste is at a certain level. As a point of reference, the amount of ions present in the average tap water in Japan is 10–50 μS/cm. For ion removal from tap water, the ion exchange is the most appropriate from among the methods listed in Table 5.1. The principle and characteristics of each of the liquid waste treatment methods in the table are described in turn below.

5.3.1 Coagulation-Sedimentation (Adsorption)

In the coagulation-sedimentation, chemicals are added to the liquid wastes to form insoluble precipitates called sludge; radionuclides in the liquid wastes are removed by means such as coprecipitating them with the precipitates, forming mixed crystals with the precipitates, and adsorbing or occluding them onto the precipitates [4]. For example, metal ions in a liquid solution are adsorbed to a colloid surface with a slightly negative charge, which causes the metal ions to precipitate in the form of large particles.

The coagulation-sedimentation process normally consists of the following four steps: (1) addition of chemicals and pH adjusters to form precipitates; (2) coagulation; (3) sedimentation; and (4) solid-liquid separation.

In the first step, the liquid waste that contains added chemicals is stirred sufficiently so that the chemicals are mixed in and sedimentation occurs throughout the liquid waste. While increased stirring time enhances adsorption of radionuclides to precipitations, it may also increase formation of colloids, which do not readily precipitate. For this reason, a laboratory experiment should be conducted in advance in order to determine an appropriate length of stirring time. In the second step, the liquid waste should be stirred gently in order to promote coagulation of the produced precipitates and to form large aggregations of precipitates that can be readily settled out. For the sedimentation (step 3) and solid-liquid separation (step 4) of the formed sludge, spontaneous precipitation, centrifugal separation or filtration is used. The water content of dehydrated sludge is approximately 50–80 %, and is thus high. Burial disposal of this sludge requires solidification using materials such as cement.

Table 5.2 shows the pH values that are appropriate for precipitation formation and the expected values of the decontamination factor (DF). These values are

Table 5.2 Coagulating agents that are used for the removal of major radionuclides [4]

Nuclide	Coagulating agent	pH	DF
^{90}Sr	Calcium carbonate[a]	10.5	>100
	Calcium phosphate[b]	>11	>100
	Barium sulfate[c]	≥8.5	>100
	Ferric hydroxide[d]	7–13	Depends on pH
^{137}Cs	Ferrocyanides[e]	6–10	>100
^{106}Ru	Ferric hydroxide[d]	5–8.5	5–10
	Cobalt sulfide	1–8.5	30–1,000
Rare-earth elements such as ^{95}Zr, ^{95}Nb and ^{144}Ce	Ferric hydroxide[d]	≥8.5	100–1,000
	Aluminum hydroxide[d]		
Pu, Am	Ferric hydroxide[d]	7–12	>1,000

Precipitation reactions:

[a] $\mathrm{Ca(OH)_2 + Na_2CO_3 \rightarrow CaCO_3 \downarrow + 2NaOH}$

[b] $\mathrm{3CaCl_2 + 2Na_3PO_4 \rightarrow Ca_3(PO_4)_2 \downarrow + 6NaCl}$

[c] $\mathrm{Ba^{2+} + SO_4{}^{2-} \rightarrow BaSO_4 \downarrow}$

[d] $\mathrm{M}^{n+} + n\mathrm{NaOH} \rightarrow \mathrm{M(OH)}n \downarrow + n\mathrm{Na}^{+}(\mathrm{M:Fe,Al})$

[e] $\mathrm{2MSO_4 + K_4Fe(CN)_6 \rightarrow M_2Fe(CN)_6 \downarrow + 2K_2SO_4(M:\ Ni,\ Cu)}$

given for some examples of coagulating agents that are used for the major radionuclides [4]. When liquid wastes that contain multiple radionuclides are treated, more than two kinds of coagulating agents are combined and used. The most appropriate amount of coagulating agents to be added is approximately 100 ppm. Excessive addition of coagulating agents causes an unnecessary increase in the amount of sludge to be formed.

The advantages of the coagulation-sedimentation are that: it is well-suited for treatment of a large volume of liquid waste; it is not susceptible to effects from ion and salt concentrations in the liquid waste; and its treatment cost is low. On the other hand, the disadvantages of the coagulation-sedimentation are as follows: its DF values are low at approximately 10–100; it results in formation of a large amount of sludge; and operation of equipment required for this method calls for relatively high skills.

The DF is generally defined as the ratio between the pre-treatment radioactivity concentration and the post-treatment radioactivity concentration; the higher the value of DF, the more desirable. In the case of the evaporation, which is described in Sect. 5.3.3, DF is defined as the ratio between the radioactivity concentration of the original liquid waste and that of the concentrated liquid waste.

5.3.2 Filtration

Filtration is a solid-liquid separation process which separates the liquid components and insoluble solid components from each other by allowing the original liquid waste to contact a filtering medium. The available filters include precoat filters, which use filter aids; membrane filters, which use no filter aids and produce only a small amount of filter sludge; and mechanical filters such as electromagnetic filters. Filter aids are substances that are placed upstream from a filter to avoid filter clogging and capture fine particles. Diatomaceous earth is a typical filter aid.

Prior to the use of a precoat filter, a filter aid is deposited on a mesh installed in a filter, which is called an element. Then, liquid waste is passed through the precoat filter, and insoluble solid components are captured by the filter aid. When the filter is clogged and the differential pressure reaches a certain value, the captured insoluble solid compounds and the filter aids are retrieved by backwashing. As a result, filter aids is retrieved as a secondary waste; sometimes the amount is more than 10 times the volume of the captured insoluble solid compounds, and this secondary waste generation is a weakness of the filtration.

There are various styles of membrane filters available. They are classified according to their driving force to purify and to concentrate the solution. Membrane filters utilize: (1) pressure gradient, (2) concentration gradient, (3) electrical potential gradient, and (4) temperature gradient. Of the membrane filters, those that utilize the pressure gradient as their driving force for filtration are the most common. Based on the size of the particulate substances in the liquid waste to be filtered, the filtration is classified into the microfiltration, ultrafiltration, nanofiltration and reverse osmosis. The main characteristics of these methods are shown in Table 5.3.

Table 5.3 Filtration methods that utilize the pressure gradient as the driving force for filtration [5]

Filtration method	Physical structure of membrane	Separation method	Particles to be removed	Operating pressure
Microfiltration	Symmetric membrane	Sieving effect	A few 100 nm to 10 μm	50–500 kPa
Ultrafiltration	Asymmetric membrane	Sieving effect	A few nm to a few 100 nm	<1.4 MPa
Nanofiltration	Asymmetric membrane	Molecular diffusion	A few tenths of nm to a few tens of nm	<4 MPa
Reverse osmosis	Asymmetric membrane	Molecular diffusion	A few tenths of nm to a few tens of nm	>5–10 MPa

In the microfiltration and ultrafiltration, the solid contents are removed by the physical sieving effect induced when liquid waste constituents pass through the fine pores on a filter. Although both methods rely on the same principle, i.e., the sieving effect, they differ in the structures of the membranes that are used and the size of particles to be removed. Microfiltration utilizes a symmetric membrane, the entire cross section of which consists of a uniform layer. In contrast, ultrafiltration utilizes an asymmetric membrane made of a uniform material consisting of a thin, highly dense layer on the primary side and a spongy support layer on the secondary side. The highly dense layer functions as a filter medium in this asymmetric membrane.

In both the nanofiltration and reverse osmosis, asymmetric membranes are used; the process of separation relies more on the difference in the extent of diffusion than on the physical size of the constituents of the liquid. While water molecules can easily pass through nanofilters, impurities other than water—e.g., hydrated large ions, which are large molecules and cannot readily diffuse, and salts—do not readily pass through nanofilters. Although the principle and the structure of the membrane used for the nanofiltration is the same as those of the reverse osmosis, the separation performance of the nanofiltration is low. Regarding reverse osmosis, it is a process to separate the solvent and the solute from each other using the phenomenon that the solvent (usually water) moves to the low concentration side of a reverse osmotic membrane (semipermeable membrane) when pressure higher than the osmotic pressure is added to the high concentration side; the direction of the movement of the solvent in reverse osmosis is the opposite from that of osmosis.

The choice of the above-mentioned filtration methods depends on the kind of constituent to be removed with filtration. It is considered that clogging of minute filter pores can be suppressed by selecting the pore size according to the size of the constituent to be removed. However, if the selected pore size is small relative to the size of the constituent, the pores are susceptible to clogging. Furthermore, in this case, the filtration area necessary to secure the desired throughput increases and high pressure needs to be applied, which calls for the use of large equipment. Therefore, selection of appropriate pore size is important so as to reduce clogging and prevent the required pressure from becoming too high.

The peak values of the diameters of the crud particles in the liquid waste from a BWR plant occur at 1–10 μm. There are few crud particles with diameters of 0.1 μm or less. For this reason, the pore size of 0.1 μm or less is used for hollow fiber membranes, which is one type of membrane filter used for filtration at BWR plants.

Here, hollow fiber membranes refer to membrane filters which have been processed to create hollow fibers of 0.5–30 mm in diameter. Hollow fiber membranes are made of polymers such as polyethylene and polyolefin and correspond to microfiltration, ultrafiltration or reverse osmosis membranes shown in Table 5.3. Hollow fiber membranes, which provide large filtration area, are used widely for household water and industrial applications and also at water filtration plants. While many of the hollow fiber membranes filter liquid as it passes through the fiber from the external to internal surfaces, a reverse type is also available. For actual filtration equipment, numerous hollow fiber membranes are bundled to form a single module, which is installed as a filter element.

As with ordinary filters, it may be possible to perform filtration with membrane filters by passing the entire volume of liquid through the filters. However, because the pore size of the membrane filters is small, the membranes can be clogged by fine particles and impurities within a short time. In order to avoid adhesion of fine particles and impurities to the membrane surfaces, the original liquid waste is continuously flushed in a certain direction along the membrane surfaces. As for the water in which fine particles and impurities have been concentrated (called condensed water), it is continuously discharged or sent back to the upstream side of the filter. The flow along the membrane surfaces is called cross flow, and the use of filtration membranes with this flow is called the cross-flow process. This process is commonly used for filtration with membrane filters. In this filtration process, the flow direction of the original liquid waste to be treated differs from the direction of the fine pores on the filter. Therefore, clogging of the fine pores is less likely to occur. Even when the pores are clogged, as long as the filtration membrane can be backwashed, the clogging can be removed by flushing liquid backward from the filtrate side to the liquid waste side. Thus, the cross-flow process excels in durability.

5.3.3 Evaporation [6]

In the evaporation, liquid waste is sent into an evaporator to be heated, the liquid waste volume is reduced through the removal of the volatile constituents such as water, and the constituents containing non-volatile radionuclides are concentrated. The volatile constituents such as water vapor are turned into condensate. This method is superior to other liquid waste treatment methods in terms of decontamination and volume reduction. In evaporative treatment of liquid waste that contains non-volatile radionuclides, the values of 10^3–10^4 and approximately 100 are achieved for the DF and the volume reduction factor, respectively. This method is used extensively as it is well-suited to treat, in particular, liquid waste containing

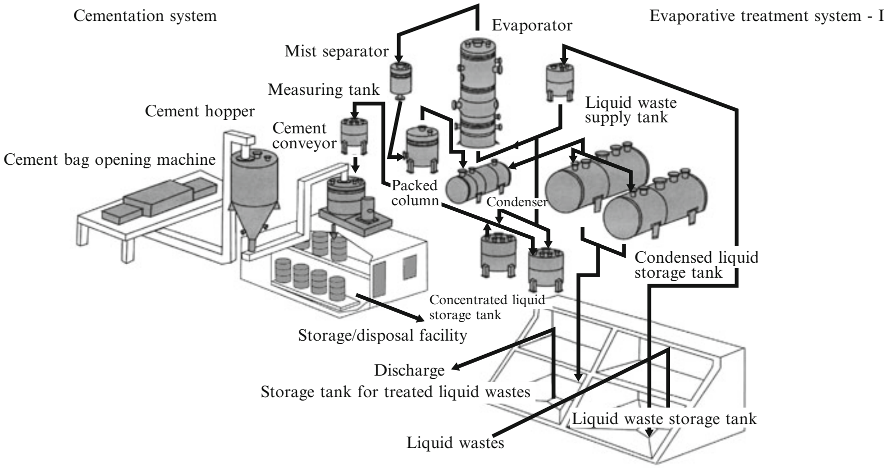

Fig. 5.1 Example of immersion evaporative concentrator and cementation system based on the out-drum mixing

a large amount of insoluble solids and ions that cannot be treated by the ion exchange. On the other hand, the treatment cost of the evaporation is high and the treatment capacity of this method is generally small, both of which are weaknesses of the method.

Fundamentally, the evaporator consists of equipment which transfers heat to the liquid and equipment which separates vapor and liquid phases from each other. Moreover, there are two types of evaporators: immersion (natural circulation) evaporators and forced circulation evaporators. In an immersion evaporator, the liquid waste is pool boiled (pool boiling results from heating of the liquid underneath its free surface) outside the heating tubes of the evaporator and undergoes natural convection in order to concentrate the waste by evaporation. In contrast, in a forced circulation evaporator, the liquid waste is put through forced convection within the heating tubes. For this reason, the concentration factor and the decontamination efficiency of forced circulation evaporators are higher than those of immersion evaporators, and thus are effective at concentrating effervescent liquid waste. As an example of an immersion evaporative treatment system, Fig. 5.1 shows an evaporation facility operated by the Nuclear Science Research Institute, Japan Atomic Energy Agency (JAEA). In the JAEA facility, the evaporator is operated in connection with the cementation process.

5.3.4 Ion Exchange [7]

In the ion exchange process, ions dissolved in the liquid waste are removed with ion exchangers. Although this process is not well-suited for liquid wastes with high concentrations of dissolved ions, it is well-suited for treatment of liquid wastes with small amounts of dissolved ions. Generally, the DF of this process is large: 10^2–10^4. No high skills are required in order to operate the equipment for this process. On the other hand, the resin used for this process is expensive, and the resin regeneration procedure produces recycled liquid waste which contains radionuclides in high concentration. Furthermore, volume reduction of the spent resin is difficult, which is another weakness of this process.

As ion exchangers, synthetic organic ion exchange resins and inorganic ion exchangers are available. Synthetic organic ion exchange resins consist of a network substrate which is linked to functional groups that contain positively charged ions such as H^+ (cation exchange resins) and negatively charged ions such as OH^- (anion exchange resins). In contrast, inorganic ion exchangers are made of clay minerals such as vermiculite and montmorillonite.

Because the DFs of synthetic organic ion exchange resins are larger than those of inorganic ion exchangers, the former are generally used at nuclear facilities. Synthetic organic ion exchange resins can be classified according to the type of functional groups that constitute the resin: strong acid, strong base, weak acid and weak base. An appropriate resin is selected depending on the purpose of application. Table 5.4 shows the values of the acid dissociation constant for the functional groups that are commonly used in synthetic organic ion exchange resins. Because the ion exchange groups on the strong base ion exchange resins dissociate both in base and acid solutions, the resins can be used to ion exchange neutral salts, such as NaCl, and basic substances. Similarly, the ion exchange groups on the strong acid ion exchange resins dissociate in base solutions, thus can also be used to ion exchange neutral salts. As for weak acid and weak base ion exchange resins, they cannot ion exchange neutral salts; therefore, application of these resins is limited. However, these types of resins can be more readily regenerated than the strong acid and strong base ion exchange resins.

Treatment processes that rely on the use of ion exchange resins are: (1) the single-bed type that uses only cation exchange resins, (2) the double-bed type in which the cation and anion exchange resins are placed in series for use, and (3) the mixed bed type in which a mixture of the cation and anion exchange resins are used. In order to remove both cations and anions from a solution, either the double-bed or the mixed bed type needs to be used. For example, when a CsCl solution is treated, the following ion exchange reaction occurs.

Table 5.4 Acid dissociation constant of functional groups that are commonly found in organic ion exchange resins [7]

Cation exchange resin		Anion exchange resin	
Functional group	p*K*	Functional group	p*K*
$-SO_3H$ (strongly acidic)	1–2	$\equiv N^+$(strongly basic)	1–2
$-PO_3H_2$	2–5	$=N$	4–6
$-COOH$	4–6	$=NH$	6–8
$-OH$ (weakly acidic)	9–10	$-NH_2$(weakly basic)	8–10

$pK = -\log$ [Acid dissociation constant]

Table 5.5 Removal efficiency of ion exchange resins [4]

	Single – bed method		
Nuclide	Cation exchange resin	Anion exchange resin	Mixed – bed method
^{91}Y	86–93	94.2–98.5	97.6–98.7
^{89}Sr	99.1–99.8	5–7	99.95–99.97
^{140}Ba-^{140}La	98.3–99.0	36–42	99.5–99.6
^{137}Cs	99.8	9	99.8
^{95}Zr-^{95}Nb	58–75	96.4–99.9	90.9–99.4

$$\begin{aligned} &R - H + Cs^{+} \leftrightarrow R - Cs + H^{+} \\ &R^{1} - OH + Cl^{-} \leftrightarrow R^{1} - Cl + OH^{-} \\ &2H^{+} + OH^{-} = H_2O \end{aligned}$$

Because H_2O is weakly dissociated, the ion exchange reaction proceeds to the right in the above equations, removing Cs^{+} and Cl^{-}. Here, R and R^{1} represent the resin matrices of cation and anion exchange resins, respectively.

Since metal ions are cations, they are removed with the use of cation exchange resins as for Cs^{+} in the above equation. If metal ions, for example, are converted to anion complexes through complex formation reactions and become stable as anions, the metal ions can be removed by anion exchange resins.

The DF of ion exchange depends on the ion exchange group in the ion exchange resins to be used. However, the value of the DF decreases in the order of the mixed bed type, double-bed type and single-bed type. Table 5.5 shows the DFs of selected ion exchange resins [5]. If the concentration of ions in the water to be treated is approximately the same as that in ordinary natural water or tap water, the following trends are known for the power of ion exchange resins to exchange and fix ions.

(1) Strong acid cation exchange resins:

$$Th^{4+} > La^{3+} > Ce^{3+} > Sr^{2+} > Ca^{2+} > Co^{2+} > Mg^{2+} > Cs^{+} > K^{+} > Na^{+} > H^{+} > Li^{+}$$

(2) Weak base anion exchange resins:

$$SO_4{}^{2-} > C_2O_4{}^{2-}(\text{oxalate}) > I^{-} > NO_{3^{-}} > CrO_4{}^{2-} > Br^{-} > Cl^{-} > CH_3COO^{-}(\text{acetate}) > F^{-}$$

Accordingly, the selectivity of ions by strong acid cation exchange resins increases with the increasing value of the valence of an ion. Among the ions with the same valence, the selectivity increases with the increasing atomic number.

5.4 Solid Waste Treatment

5.4.1 Incineration [8]

Solid radioactive materials including those generated by the operation and maintenance of facilities are usually treated after they are sorted into flammable, flame-resistant (e.g., vinyl chloride and rubbers), and non-flammable materials (e.g., insulation materials such as rock wool, metal and glass). For the treatment of flammable materials, incineration is widely used, which allows large reduction in the volume of the materials and yields chemically stable residue (incineration ash).

As for incineration, the excess air combustion and the suppressed combustion are available. In the former process, the amount of combustion air that is larger than the theoretical combustion air (the excess air ratio of 1 or larger) is supplied for combustion. In the latter process, the excess air ratio of less than 1 is used for combustion. If the percentage of the volume of oxygen in the air is assumed to be 21 %, the theoretical combustion air necessary for burning waste (the volume of air required for complete combustion of the waste) can be evaluated by the following equation.

$$L_0 = \left(\frac{c}{12} + \frac{h}{2} + \frac{s}{32} - \frac{o}{32}\right) \cdot \frac{22.4}{0.21} \tag{5.5}$$

L_0: the theoretical combustion air [$\mathrm{Nm^3}$/kg-waste]
c, h, s, o: the respective mass of carbon, hydrogen, sulfur and oxygen per unit mass of waste [kg/kg-waste]

In the actual waste combustion, the theoretical combustion air is insufficient for complete combustion, thus unburned gas and soot are generated. Therefore, the use of the excess air combustion, which requires attention be paid to the excess air ratio (λ_a), may be desirable. The values of the excess air ratio vary according to the quality of the waste and the performance as well as the type of the incinerator and range from approximately 1.3 to 2.0. If the quality of the waste is high, that is, the waste consists of few flame-resistant materials and materials with low water content, a small value can be chosen for λ_a. However, if the quality of the waste is low, a large value needs to be chosen for λ_a; as a result, the amount of combustion gas that requires treatment increases. The amount of wet combustion gas, which is generated by incineration and contains water vapor, can be calculated from the following equation:

$$
\begin{aligned}
V_w = {} & \underbrace{22.4\left(\frac{c}{12}\right)}_{\text{CO}_2\ \text{generated}} + \underbrace{22.4\left(\frac{h}{2}+\frac{W}{18}\right)}_{\text{H}_2\text{O generated}} + \underbrace{22.4\left(\frac{s}{32}\right)}_{\text{SO}_2\ \text{generated}} + \underbrace{0.21(\lambda_a - 1)L_0}_{\text{Excess O}_2} \\
& + \underbrace{0.79\lambda_a \cdot L_0 + 22.4\left(\frac{n}{28}\right)}_{\text{N}_2\ \text{from the air} \ + \ \text{N}_2\ \text{generated}} \\
= {} & 1.867c + 11.2h + 1.244W + 0.7s + 0.8n + (\lambda_a - 0.21) \cdot L_0 \qquad (5.6)
\end{aligned}
$$

where V_w is the amount of wet combustion gas [Nm^3/ kg-waste] and n, W is the respective mass of nitrogen and water per unit mass of waste [kg/kg-waste].

In the suppressed combustion, in which waste is burned using an amount of air less than the theoretical combustion air, primary and secondary combustion chambers are usually installed. In the primary combustion chamber, flammables are gasified with insufficient oxygen. In the secondary combustion chamber, the pyrolysis gas generated in the primary combustion chamber is burned with excess oxygen. This method is well-suited for incineration of wastes such as plastics that can easily generate soot. However, the use of this method requires attention to prevent tar formation and explosions. Furthermore, because the ashes generated from the application of this method contain carbon, the method is not highly efficient in reducing the weight and volume of the original waste.

Table 5.6 shows example compositions of the solid wastes that were taken into consideration for designing excess air combustion incineration facilities for the treatment of flammable wastes and suppressed combustion incineration facilities for the treatment of flame-resistant wastes. When polymer wastes such as plastics

Table 5.6 Example compositions of wastes to be treated by the excess air combustion method and the suppressed combustion method [9, 10]

Main materials to be treated	Flammable wastes	Flame – resistant wastes
Combustion method	Excess air combustion	Suppressed combustion
Constituents of assumed wastes	Cloth: 15–30 %	Cellulosic materials: 10 %
	Paper and wood: 40–50 %	
		Polyethylene: 40 %
	Plastic (polyethylene, polypropylene, rubber) : 20–30 %	Rubber and related materials: 30 %
		Vinyl chloride: 20 %
	Vinyl chloride : up to 3 %	Water content: 10 %
		Ash content: 10 %
Calorific value of wastes	15–25,000 kJ/kg (Range of calorific values of wastes generated at nuclear power plants)	23,800 kJ/kg

and vinyl chloride are burned, the pressure fluctuations inside the incinerator may become large as a result of a rapid combustion of the pyrolysis gas. In this case, it may be difficult to confine radioactive materials (negative pressure control). Therefore, the use of the suppressed combustion is effective for the incineration of waste that contains a large amount of polymer waste.

Incineration systems commonly used for treating radioactive solid wastes consist of a waste supply unit, an incinerator, an incineration ash recovery unit, off-gas treatment equipment and other components. For designing and managing the operation of incineration equipment, treatment of off-gas generated from incineration becomes important. At many of the incineration facilities belonging to current nuclear facilities in Japan, off-gas generated from incineration is treated by ceramic filters made of silicon carbide (SiC), HEPA and other particulate cleaning systems to remove particulate matters that contain radioactive materials. After confirming that the off-gas concentration is at or lower than that set forth by the release limit, the off-gas is released through the stack. Because volatile radionuclides such as ^{3}H, ^{14}C and ^{129}I cannot be removed by particulate cleaning systems, scrubbers are in some cases added to the incineration system, which allows water to absorb these volatile radionuclides for recovery.

Figure 5.2 shows, as an example of incineration facilities that utilize the excess air combustion, an incineration facility operated by the Nuclear Science Research Institute at JAEA. At this facility, a vertical type incinerator is used; the system is designed in such a way that the constituents that could not be completely combusted in the incinerator are combusted on the surface of ceramic filters maintained at a high temperature (the primary and secondary ceramic filters in the figure). Furthermore, after the off-gas of high temperature is passed through a heat

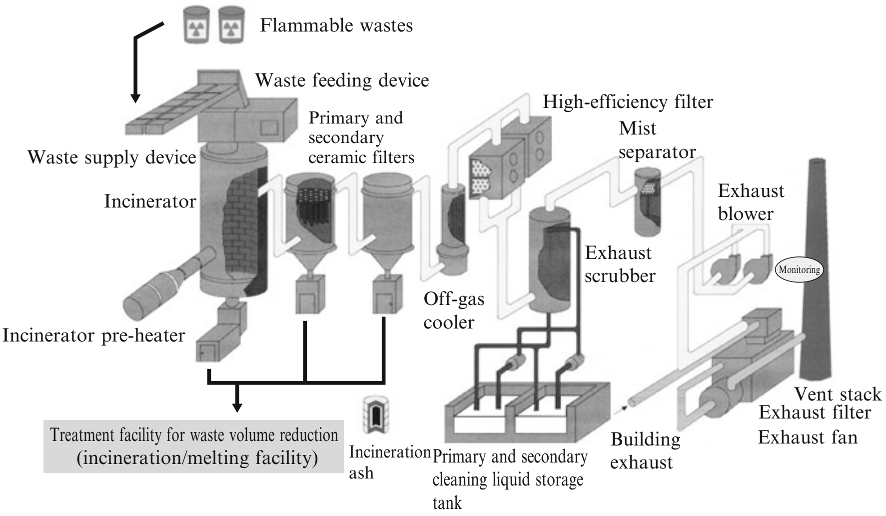

Fig. 5.2 Example of an incineration facility that uses the excess air combustion method

exchanger (off-gas cooler) and cooled down to 200 °C, fine particles are removed by HEPAs designed for a high temperature environment.

At nuclear power plants in Japan, high temperature incineration furnaces, which are second generation incinerators and capable of burning flame-resistant materials, have been introduced. In high temperature incineration furnaces, non-flammable materials such as small metal pieces, insulation materials such as rock wool, and concrete pieces and flammable materials are ground and mixed; the mixture is burned with supplementary fuel at high temperatures of 1,400–1,500 °C, and the resulting ashes are melted. The melt is dropped into water, turning it into glassy granules (porous granulated pieces approximately 3 mm in size). The volume reduction factors of light metals, air filters and insulation materials are 1/5 to 1/3 while those of other flammable materials are approximately 1/100. Granules contain silicon, iron, aluminum and oxides of other metals. The bulk specific gravity of granules is larger than that of ashes formed in conventional incinerators. Furthermore, the water leaching rate of granules is small, and granules are not easily scattered. Finally, because granules are characterized by a certain degree of flowability, they are easy to handle and can be solidified more readily than regular incineration ashes.

In high temperature incineration furnaces, most of the solid wastes generated at nuclear facilities can be treated. Therefore, the screening and sorting process required for incineration can be simplified considerably, which, as a result, makes it possible to save labor and reduce radiation exposure to workers. According to past records of demonstration plant operation, high temperature furnaces are able to treat wastes characterized by a ratio of non-flammable to flammable materials in the range of 80/20 to 20/80 at a throughput of 100 kg/h.

Regarding operation management of high temperature incineration furnaces, when the waste composition deviates from the prescribed composition, the system may need to be shut down. Therefore, adequate management of waste composition is even more important for high temperature incineration furnaces than for conventional incinerators; this calls for information on the composition ratio of the waste to be treated and the waste classification process.

5.4.2 Compaction

The objective of this treatment method is to reduce the volume of compactable solid waste. Reduction of waste volume is effective in that it creates sufficient storage space within facilities that retain waste and also brings down the number of waste forms to be treated, thus lowering the disposal cost. Commonly, compressive force of approximately 50 kN to 3 MN (N: Newton; 1 N is approximately 0.10 kgf) is used. Although it varies according to the material, the volume reduction factor can be as large as approximately 5. On the other hand, there is a high compaction that allows attainment of approximately 10 for the maximum volume reduction factor with the compressive force of approximately 10–20 MN. Two methods are available for compacting the waste: a method in which the waste is compacted directly in drums, and a method in which the waste is compacted inside pressing chambers. Furthermore, depending on the direction of compaction, the compaction can be classified into uniaxial, biaxial and triaxial compaction. When drums filled with waste as a whole are put through the compaction process for treatment, they are squeezed vertically, in some methods, to reduce the diameter to store the waste in another drum. The method to compact waste in drums is a uniaxial compaction, and the required equipment is simplest. However, the use of this method imposes limitation on the size of the waste to be compacted and may cause damage to the inner walls of drums when hard objects such as metals are compacted. If a pressing chamber is established for compaction treatment, a multiaxial method can be applied. Because the volume of the pressing chamber can be set large, it is possible in this approach to increase the amount of waste treatable within a single compaction session and also to treat large-sized wastes.

The volume reduction factor varies according to the objects to be compacted. However, except for the case of metals, no further volume reduction can be expected from application of 10 MN (approximately 1,000 tons) or more in pressure. In the case of metals, the volume reduction factor does not increase significantly with the application of 10 MN or more in pressure. However, when a pressure of 16 MN or higher is applied to metals, a small increase in the volume reduction factor can be confirmed. It should be noted that compaction cannot reduce the volume of waste down to the value close to the true density of the constituents of the waste. For the case of those metals characterized by the smallest volume reduction factors, even with high pressure treatment at 16 MN, they can be compacted down to only approximately 40–50 % of the true density. A larger volume reduction factor can be achieved for low density plastic and related materials. Nonetheless, if these materials are compacted without being stored in containers, the volume may increase after cessation of pressure application, that is, the spring-back phenomenon occurs, and the volume reduction factor may become low.

When waste is compacted, dust is dispersed and free water in the waste seeps out. (If the waste contains free water, it is desirable to separate the water prior to the treatment). Therefore, measures against internal exposure of workers and those for waste water treatment need to be implemented.

5.4.3 Melting

5.4.3.1 Characteristics of Melting

Recently, waste treatment equipment has been invented for melting wastes in high temperature environment. The objects to be treated include inorganic materials such as metals, concrete and incineration ashes. Melting of wastes is characterized by the following: (1) a large volume reduction of waste can be achieved: (2) solid waste can be homogenized (the properties and radioactivity distribution can be homogenized); and (3) the waste can be stabilized. These characteristics are effective for the rational treatment of radioactive waste.

(1) Waste volume reduction

In the melting, the waste can be treated so that its density becomes close to the true density of the waste constituents; therefore, the waste volume can be reduced significantly. This treatment is expected to be highly effective at waste volume reduction in particular of waste that includes numerous voids such as metal pieces of various shapes stored directly in a drum.

(2) Waste homogenization

Another characteristic of melting is that it can homogenize the distribution of radionuclides in waste forms and constituents within waste. This characteristic is important in that it allows application of representative sample analysis to waste forms in order to evaluate their radioactivity. In a typical sample analysis, a sample is obtained from melt that has been homogenized in composition; based on the measurement result of the radioactivity from the melt sample, the radioactivity of the parent melt is evaluated. The melting is an inexpensive and effective method to acquire highly reliable radioactivity information about waste from research and other facilities, where raw wastes are generated frequently in multiple lots and heterogeneous in radionuclide composition.

In the melting, waste is liquefied at high temperatures favorable for homogenization. However, in an actual melting, operation conditions such as the waste characteristics and melting temperature can sometimes affect the degree of homogenization. For this reason, operation conditions that would yield the desired degree of homogeneity need to be checked in advance. Especially if non-flammables made of various materials are simultaneously processed by melting, caution is necessary as the viscosity of the melt can change significantly according to the constituents and other characteristics of the melt.

(3) Stabilization of wastes

In the melting of waste consisting of inorganic materials, waste is melted once and then solidified; this allows stabilization of radionuclides. The solidified non-metallic materials that form as a result of the melting procedure are called slag. Because concrete and incineration ashes, which are commonly processed by the melting, contain a large amount of silica (SiO_2), the solidified form of concrete and incineration ashes becomes glass-like or rock-like. The glass-like form is made of a silica network that contains chemical elements other than silica. Thus, only small amounts of elements leach out of the glass-like form in the presence of water, making the form stable. The rock-like form has the crystal structure of the elements present in the waste. The leachability of the rock-like form is higher than that of the amorphous glass-form. Stabilization of radionuclides in waste is effective for safe waste disposal. Because of these characteristics of melting on inorganic materials, melting has been adopted for treating incineration ashes formed at refuse incineration plants in general industry. Slag generated at these plants is then reused as, for example, base materials for roads and concrete aggregate.

In the course of vitrification of high-level radioactive waste, glass melted at high temperatures and radioactive liquid waste are mixed and solidified, which allows confinement and stabilization of radionuclides. Thus, vitrification of radioactive wastes is based on the same principle as melting.

5.4.3.2 Melting

In the melting, because of such properties as the melting point of the waste to be treated, the waste needs to be melted at a high temperature, approximately 1,500 °C, and a number of heating methods have been examined. Currently, the heating methods that are used for the melting of radioactive waste include: the high frequency induction heating method, the plasma heating method, the microwave heating method, and the Joule heating method. Table 5.7 summarizes the major heating methods.

5.4.3.3 Application Examples

Melting has been introduced at some nuclear power plants and facilities of the JAEA. Figure 5.3 shows the melting system in place at the Nuclear Science Research Institute of JAEA. This system is designed for treating non-conducting, non-flammable wastes such as glass, concrete, insulation materials (e.g., glass wool and gypsum boards) and incineration ashes. Therefore, the system utilizes the plasma heating method as it allows heating of the waste regardless of the materials. Furthermore, the off-gas from the melting system is treated by the off-gas treatment system of the incinerator that is adjacent to the melting system. In the plasma melting furnace, air is used as plasma gas, which as a result generates nitrogen oxides (NOx). For this reason, off-gas treatment system is equipped with NOx removal equipment and also with filters and other components for removing radionuclides in order to protect the environment.

5.5 Solidification

Except for gaseous and liquid wastes that are released directly into the environment, radioactive wastes generated are ultimately either solidified into forms that are adequate for burial disposal or packed into vessels such as drums to complete the waste treatment. This last treatment process is called waste form formation. Depending on the solidification method to be adopted, safety performances expected for the disposal facility for confining radionuclides and the generated

Table 5.7 Melting methods [11]

The heating method and target wastes	Process	Characteristics
Plasma melting; applicable to all materials	A plasma arc is generated from the plasma torch placed inside the incineration furnace. With the heat generated from the plasma arc, waste is melted at 1,300–1,700 °C	Because a very high temperature of 5,000 °C or higher can be easily generated, the organic constituents in waste can be thermally decomposed into gases of low molecular weight. In this method, non-volatile inorganic constituents are melted and cooled to form stable slag. Although all kinds of wastes can be melted and solidified, a significant amount of fume and dust is generated. For this reason, depending on the characteristics of the waste to be treated, an appropriate feeding method needs to be devised
Microwave melting; inappropriate for materials such as dielectrics and metals as they reflect microwaves	Microwaves of 915 or 2,450 MHz in frequency are irradiated to vibrate atoms or molecules in dielectrics. The heat generated from interatomic or intermolecular friction is used as thermal energy to melt waste. The melting temperature is 1,300–1,500 °C	This method has a proven record as a technique to melt and solidify sewage sludge and incineration ashes of general waste. Electrical discharge phenomena occur due to metals and unburned carbide. Because these phenomena can cause adverse effects such as formation of air bubbles in slag, the amount of metals and unburned carbide in waste needs to be maintained below a certain level
High frequency induction melting; applicable to conductive materials	A high frequency current is applied in the coil placed around the target to be heated, and a magnetic field is generated inside the coil. The current induced in the target is used to heat and melt the target itself. The melt temperature is 1,500–1,600 °C	This method has a track record for general industry use with metals. The heating efficiency is high as the target itself is heated directly

(continued)

Table 5.7 (continued)

The heating method and target wastes	Process	Characteristics
High frequency induction melting (also called conductive crucible method); applicable to all materials	A high frequency coil is placed around a conductive crucible. An induced current is used to heat the crucible, which melts the waste inside. As necessary, materials for vitrification (e.g., SiO_2) are added to the waste and melted at 1,000–1,300 °C	Because the waste is melted by heat transferred from the crucible, this method is not dependent on the waste characteristics. (Note that if the target to be heated itself is conductive, then heat from the induced current is also added to the waste.)
Joule heating and melting; applicable to conductive materials	Waste is placed in a crucible such as that made of ceramic or heat resistant material. Electrodes are used to pass electricity through the waste. With the Joule heat generated by the electric resistance of the waste, the waste is heated and melted	The heat source needs to be controlled according to the waste composition

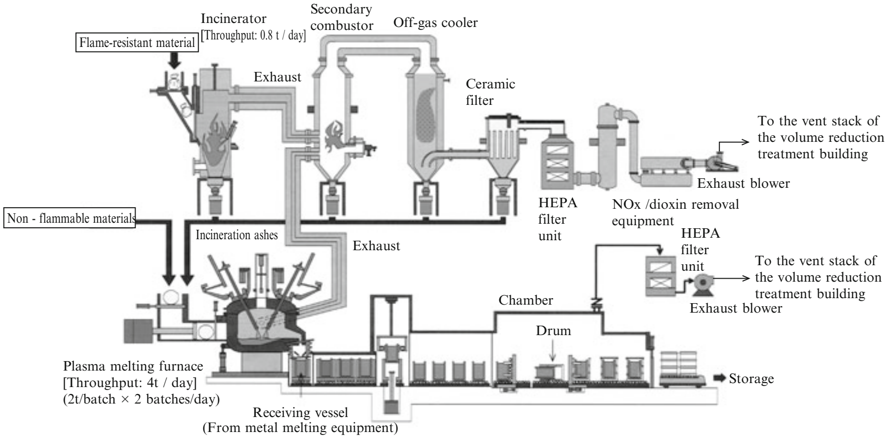

Fig. 5.3 Example of plasma melting facility

amount of waste forms to be disposed of vary. These points need to be noted when selecting the solidification method. Below, commonly used solidification methods are summarized separately for the cases of treating low-level and high-level radioactive wastes.

Table 5.8 Solidification treatment methods for low-level radioactive wastes [6]

		Cementation	Plastic solidification	Bituminization
Treatment process	Complexity	Low	Low	Low
	Flexibility	High	Average	High
	Cost	Low (3)[a]	High (1)	High (2)
Waste characteristics	Ratio of solidification material to waste	Average (3)	High (2)	High (1)
	Compressive strength	Average	Above average	Low
	Impact resistance	Average (1)	Above average (2)	Average (3)
	Fire resistance	High (3)	Below average (1)	Low (2)
	Stability against radiation	High (1)[b]	Average (3)	Average (2)
	Ability to confine radionuclides[c]	FP: low, An: high	FP: low, An: low	FP: high, An: low

[a] Numbers in parentheses indicate the descending order of highness in each characteristic
[b] Neither physical nor chemical property changes occur with the exposure dose of less than 10^8 Gy
[c] *FP* fission product, *An* actinide

5.5.1 Solidification of Low-Level Radioactive Wastes [6]

When low-level radioactive wastes such as concentrated liquid wastes and sludge formed from applications of the coagulation-sedimentation are solidified, cement is widely used as a solidification material because of the convenience in treatment and cost. Cementation has been used for more than 40 years as a method for solidifying radioactive wastes. Although concentrated liquid waste to be solidified contains water, adjustment of water contents is required in the process of solidification because cementation needs a certain amount of water. To overcome this demerit of the cementation, asphalt and plastic solidification have been put into practical use. In these methods, unwanted water content in the waste is separated from the solid constituents by evaporation; the only solid constituents in the wastes are then mixed with solidification materials. The asphalt and plastic solidification are able to reduce the volume of generated waste forms to approximately 1/3 and 1/5, respectively, of that generated from the cementation. Table 5.8 shows the characteristics of the above-mentioned solidification methods in application of treating low-level radioactive wastes.

5.5.1.1 Cementation

Because of the amendments of the Laws Concerning the Prevention from Radiation Hazards due to Radioisotopes, etc and the Act on the Regulation of Nuclear Source

Material, Nuclear Fuel Material and Reactors in 1996, sea dumping of radioactive waste is now prohibited. Back when sea dumping was assumed for use as a disposal method, it was required that the waste forms disposed of in the sea would not be destroyed by water pressure and would definitely land on the ocean floor at a specified depth (more than 4,000 m deep). Thus, waste to be disposed of in the sea was required to be cemented in such a way that the specific gravity of the cemented waste would be 1.2 or larger.

Because the prohibition against the ocean disposal came into effect and land burial disposal started to be presumed, the guidelines for cementing low-level radioactive waste have been replaced by those having burial disposal in mind. Currently, the technical standards required for cemented forms for burial disposal are stipulated in the "Notification Specifying the Technical Details Concerning Burial Operation of Nuclear Fuel Material, etc., No. 2" (hereafter Technical Notice No. 2), which calls for the use of JIS R 5210 (Portland cement), JIS R 5211 (blast-furnace cement), or cement with quality equal to or higher than that of JIS R 5210 and JIS R 5211. The technical standards are described in Sect. 5.5.3.

The main constituents of Portland cement are SiO_2, calcium oxide (CaO) and alumina (Al_2O_3). By adjusting the blending ratio of these main constituents, various types of Portland cement have been made available. Furthermore, in addition to ordinary Portland cement, some of the Portland cement types that are prescribed by JIS R 5210 are high-early-strength Portland cement, ultra-high-early-strength Portland cement, moderate-heat Portland cement, low-heat Portland cement, sulfate-resistant Portland cement and low-alkali Portland cement. Depending on the application, a certain type of Portland cement is utilized.

On the other hand, blast-furnace cement is made by mixing finely powdered blast-furnace slag produced at steel mills into the cements. The long-term strength of blast-furnace cement is enhanced so that it becomes larger than that of Portland cement types, and the sea water resistance and chemical resistance of blast-furnace cement are excellent. At the same time, because the cement solidifies slowly compared to Portland cement, its initial curing is more important.

In-drum and out-drum mixing are available as cementation. For the in-drum mixing, the waste to be solidified and cement are mixed inside a drum or other containers for solidification. For the out-drum (in-line) mixing, after the waste and cement are mixed in a mixing device, the mixture is poured into a drum or other containers. The equipment for the in-drum mixing is simpler than that for the out-drum mixing. Moreover, the cost and the maintenance required for the in-drum mixing are lower and easier, respectively, than those required for the out-drum mixing; these are some of the advantages of the in-drum mixing. On the other hand, the treatable amount of waste at a time is small for the in-drum mixing. Because the waste and cement are mixed in a drum or other containers, the amount to be mixed needs to be restricted so that the waste-cement mixture will not spill out of the container. As a result, additional work is necessary to fill the voids formed near the top of the drum or other containers; this is a disadvantage of the in-drum mixing. Therefore, at plants at which a large amount

Table 5.9 Chemical compatibility between cement and type of wastes and measures to improve compatibility [6]

Type of wastes	Chemical compatibility	Measures
Organic ion exchange resin	Poor	Add calcium hydroxide
Coagulation – sedimentation sludge	Good	–
Boric acid waste solution	Poor	Add sodium silicate
Sulfate waste solution	Moderate	Use sulfate -resistant cement
Nitrate waste solution	Good	–
Phosphate waste solution	Good	–
Waste solution containing cleaning agent	Poor	Add anti-foaming agent
Liquid waste containing complexing agent	Poor	–
Oil, organic liquid waste	Poor	Add emulsifier
Acidic drainage water	Poor	Neutralization

of waste undergoes the cementation treatment, the out-drum mixing is used. An example cementation system based on the out-drum mixing is shown in Fig. 5.1.

The water-to-cement ratio most influences the strength, chemical resistance, water permeability and other characteristics of cemented forms. As described earlier, the waste itself contains water, and for reasons associated with workability, more water is necessary than the amount required for cement hydration; thus, the amount of water to be used for mixing the cement and waste cannot be readily adjusted. When the water-to-cement ratio becomes large, voids occur after solidification, which sometimes prevents achievement of the optimal characteristics of the cemented form.

Depending on the waste to be solidified, its constituents impede the cement hydration reaction. Therefore, ordinary Portland cement cannot be used in some cases. Table 5.9 shows the chemical compatibility of each waste type and cement as well as measures to be used when compatibility is poor.

When inhomogeneous wastes such as metal wastes and compressed wastes are cemented in a container, highly fluid cement mortar is used in order to minimize the voids in-between the waste components and between the waste and the container.

5.5.1.2 Bituminization

Asphalt, also called bitumen, is also used as solidification material for the solidification of radioactive wastes. In the bituminization, liquid or slurry waste is mixed with melted asphalt while being heated; this process removes the water contained in the waste. After the remaining solid constituent is dispersed homogeneously within the asphalt, the mixture is naturally cooled within a container to become a solid form. The technical standards required for bituminized radioactive waste to be disposed of are stipulated by the Technical Notice No. 2, which states that (1) asphalt with the penetration grade of 100 or less or asphalt with quality equivalent to or higher than JIS K 2207 needs to be utilized and (2) the mix proportion needs to be set in such a way that the proportion of asphalt is 50 % or more in weight.

The following can be considered as the advantages of bituminized radioactive waste. Asphalt itself is water-insoluble and highly resistant to water diffusion, which as a result makes bituminized radioactive wastes highly resistant to leaching to water. A large amount of solid in the waste that remains after the evaporation of water can be incorporated into bituminized waste forms, and a larger volume reduction factor can be achieved by forming bituminized waste rather than other waste forms. This property leads to a reduction of the number of waste forms. Furthermore, the plasticity and rheological (fluid flow) characteristics of asphalt are excellent. Therefore, even when pressure is exerted inhomogeneously on bituminized waste forms underground after their burial, it is expected that the waste forms will not be subjected to mechanical fracture. The cost of the bituminization itself is relatively low.

On the other hand, a disadvantage of bituminized waste forms is that they are flammable. Bituminized waste can ignite at a temperature of 290–350 °C or higher although it depends on the type of asphalt used for solidification. Furthermore, bituminized waste can ignite by reacting with oxidants such as sodium nitrate. Such chemical reactivity is the most problematic issue for the use of bituminization. In reality, fire accidents have broken out in multiple countries including Japan when bituminized waste forms were being transferred into drums. All these cases occurred while applying the bituminization process to concentrated liquid waste that contained nitrate at reprocessing plants. Another disadvantage of bituminized waste is that it is less resistant to radiation than cement-solidified waste. Therefore, with the presence of radiation, the above-mentioned excellent characteristics of bituminized waste deteriorate. Because of the low radiation resistance, application of the bituminization is limited to low-level radioactive wastes.

5.5.1.3 Plastic Solidification

The solidification material used for plastic solidification is a resin called a thermosetting resin. Compared with thermoplastic resins, thermosetting resins can be treated at lower temperature. Furthermore, among thermosetting resins, unsaturated polyester resin is commonly used. Unsaturated polyester resin is a liquid mixture of styrene monomers and unsaturated polyesters that contain unsaturated groups such as maleic acids, phthalic acids and propylene glycol polymers. By adding a hardening agent (initiator or accelerator) to solidification materials in which powdered waste has been dispersed homogeneously, the hardening reaction can be made to occur at room temperature. Waste elements are locked into the three-dimensional mesh network of the hardened resin and stabilized.

The technical standards required for plastic solidified waste to be buried are stipulated in the Technical Notice No. 2. According to the Technical Notice, it is required that the blending proportion of the plastic be 30 % or greater in weight and the hardness value (measured by the durometer method stipulated in JIS K 7215) of the produced solid form be larger than 25.

The plastic solidification has been developed and put into practical use in general industry. Although this method was once introduced into the nuclear energy field, too, it is hard to find advantages of this method over the cementation and the bituminization; thus, plastic solidification is not commonly used in the nuclear energy field.

5.5.2 Solidification of High-Level Radioactive Wastes [12]

Vitrification has been put into practical use as a treatment method to solidify high-level radioactive liquid wastes. This is attributable to the fact that glass excels in its thermal stability, leaching resistance and radiation resistance and that existing technologies from the glass industry can be applied in this treatment method.

Currently, borosilicate glass, which is made of silicon (Si) and boron (B) is most commonly used for vitrification worldwide and it has been used in Japan. Borosilicate glass is a network of glass-forming chemical elements such as Si, B and O. Some other elements such as Cs and Sr, which are waste elements in the same category as Na, Li and Ca, are situated within the network as network-modifier ions. Solidification of waste with the use of phosphate glass that consists primarily of phosphorus (P) has been examined. However, because the mixture of the waste and phosphate glass exhibits a highly corrosive nature during the melting procedure and the thermal stability of the solid form is poor, this method has not been put into practical use to create solid forms of high-level radioactive liquid wastes.

As industrial-scale technologies of continuous glass melting, the metal melter system (Atelier Vitrification de Marcoule (AVM) system) and the direct electrification ceramic melter system (Liquid Fed Joule-heated Ceramic Melter (LFCM) system) are the most common. In the former system, the glass melting furnace is heated from the outside using high frequency electromagnetic waves. In the latter system, electricity is supplied directly to the glass inside the melting furnace made of refractory bricks, and the glass itself is heated. Japan Nuclear Fuel Ltd. has adopted the LFCM system for the reprocessing plant located in Rokkasho Village, Aomori Prefecture.

Because heat is provided by high frequency electromagnetic waves in the AVM system, the metal melter cannot be enlarged. In order to increase the treatment capacity, a process is necessary to calcinate the liquid waste in advance. Some of the disadvantages of the AVM system are that the lifetime of the melter is short and the moving part of the melter requires frequent maintenance. The LFMC system has been widely used in the glass industry. Because the glass itself is heated in this system, heat can be effectively used for melting and a high capacity can be achieved. For these reasons, liquid waste can be supplied directly into the melter, and a series of processes consisting of evaporation, calcination and melting can be performed in the LFCM system.

After the high-level radioactive liquid waste generated in the first stage of the first cycle of the reprocessing procedure is neutralized, it is placed in the melting furnace with the glass materials Si and B. When the liquid waste and glass materials are heated for a few hours at a temperature of about 1,200 °C, the chemical elements in the liquid waste are melted into the molten glass phase. The molten glass, which has taken in the waste elements, is extruded through the nozzle located at the bottom of the melting furnace and then into a canister, a cylindrical container made of stainless steel, located below the nozzle; the molten glass solidifies in this canister.

The composition of high-level radioactive liquid wastes varies slightly according to the type of the fuel used and the reprocessing method used: 10–20 % of the vitrified waste in weight is the waste elements and 80–90 % of the vitrified waste in weight is the glass materials. According to the liquid waste composition, the characteristics of the vitrified waste differ. If the waste contains, for example, a large amount of sodium, the melting temperature of glass becomes low, and so do the corrosion resistance, radiation resistance, and water leaching resistance. Furthermore, if a large amount of molybdenum is present in the waste, the mechanical strength of the glass is weakened. In order to suppress these effects, the amount of waste in glass is restricted.

Behaviors of platinum group elements are an important issue for vitrification. Ruthenium, being a volatile element, burdens off-gas processing systems. Because palladium and rhodium can be easily separated out from molten glass, they go through phase separation inside the melting furnace and accumulate, which may cause the melting furnace to become clogged.

The fabricated vitrified forms are stored for approximately 30–50 years and cooled down while short-lifetime nuclides are decayed. It is being planned that the vitrified forms, after this time, will be disposed of in a deep geological formation (Chap. 6).

5.5.3 Waste Form Verification

From the viewpoint of ensuring safety of the workers engaged in radioactive waste burial disposal and of the general public around treatment plants, requirements that need to be met for burial disposal of low-level radioactive waste forms are set forth as technical standards in the “Regulations Concerning the Waste Burial Business for Nuclear Fuel Material or Material Contaminated with Nuclear Fuel Material ” and the “Notification Specifying the Technical Details Concerning Burial Operation of Nuclear Fuel Material, etc.” Table 5.10 shows the technical standards of radioactive waste forms for disposal that are set forth in them.

Businesses engaged in waste burial are required to go through inspections set forth by the government in order to ensure that the waste forms to be disposed of by

Table 5.10 Technical standards for waste forms

Relevant aspect stipulated in the technical standards	Type of wastes	Standard
Solidification materials	Cement	Cement types prescribed by JIS R5210 (Portland cement), JIS R5211 (blast-furnace cement), or cement types with quality equivalent to or higher than the aforementioned cement types.
	Asphalt	Petroleum asphalt with the penetration grade of 100 or less as prescribed by JIS K2207, or asphalt with quality equivalent to or higher than this petroleum asphalt.
	Plastic	Unsaturated polyester dissolved in styrene
Container	All waste forms	Metal containers prescribed by JIS Z 1,600 or containers with strength and sealing performance equivalent to or higher than the aforementioned containers.
Uniaxial compressive strength	Cemented forms	15 kg/cm^2 or higher
Mix proportion	Bituminized waste forms and plastic-solidified waste forms	Solidification materials need to be mixed with the waste in proportions of more than 50 % and more than 30 %, respectively, in weight.
Hardness value	Plastic-solidified waste forms	Hardness value measured by the method prescribed by JIS K7215 (durometer hardness test) needs to be 25 or higher.
Kneading and mixing	Radioactive liquid wastes, etc.	Solidification materials and admixtures need to be kneaded and mixed homogeneously. Alternatively, solidification materials that were kneaded and mixed homogeneously beforehand and admixtures, etc. need to be mixed homogeneously with waste.
	Solid wastes including metal wastes	Solidification materials that were kneaded and mixed homogeneously beforehand, admixtures, etc. need to be filled into the container so as to integrate them with the waste.
Hazardous voids	All waste forms	No voids that would cause collapse of the burial ground are allowed in the container in which waste was solidified. Furthermore, it is required that no voids exist that would enhance seepage of radionuclides.

(continued)

Table 5.10 (continued)

Relevant aspect stipulated in the technical standards	Type of wastes	Standard
Maximum radioactivity concentration	All waste forms	It is required that radioactivity concentration does not exceed the maximum value reported in the application form and other forms (e.g., a waste disposal license application and documents attached upon approval of the application which include a list of conditions).
Surface contamination concentration		The radioactive material density on the waste form surface shall not exceed 1/10 of :4 Bq/cm^2 for radioactive materials that emit alpha rays 40 Bq/cm^2 for radioactive materials that do not emit alpha rays
Materials that may damage the integrity		It is required that the waste does not contain the following materials that may damage the integrity of waste forms.
		Explosive materials: nitro compounds, nitrate esters, etc.
		Materials that react in an explosive manner upon coming into contact with water: metallic potassium, metallic sodium, etc.
		Explosive materials: acetone, gasoline, etc.
		Pyrophoric materials: metallic potassium, metallic sodium, yellow phosphorus, etc.
		Materials that significantly corrode waste forms: nitrate, perchloric acid, etc.
		Materials that would release a large amount of gas: metallic potassium, metallic sodium, etc. (Note: No specific material names are listed in the notice.)
Waste weight-bearing capacity upon burial disposal		The strength of the waste form needs to be high enough to withstand the load upon burial disposal.
Notable damage		Containers in which waste has been solidified are required to be free of notable damage.
Signage/indicators		Requirements are: (1) a sign identifying the radioactive waste, (2) a color band indicating the dose equivalent rate (if the rate exceeds 0.5 mSv/h) and (3) the waste form serial number be displayed on the waste form surface by selecting a location easily visible and using a method that does not allow easy erasing of the displayed writing.

Table 5.11 Example of methods for waste form verification [13]

Category to be checked	Non-destructive measurement	Production management	Sampling program
Solidification material		○	
Container		○	
Uniaxial compressive strength	(○)	○	○
Hardness value	(○)	○	○
Mix proportion		○	○
Homogeneity/uniformity		○	○
Hazardous voids		○	
Radioactivity concentration	○	○	○
Surface contamination density	○		
Materials that damage the waste form soundness		○	
Waste weight-bearing capacity upon burial disposal		○	
Notable damage	○		
Surface dose rate	○		
Sign/serial number	○		

burial satisfy the technical standards shown in Table 5.10. This inspection is called the waste form verification. For waste form verification, waste burial businesses are required to prepare documents (data) proving that the individual waste forms satisfy the technical standards. Table 5.11 shows an example of a waste form inspection method for verifying the compliance of the waste forms to be buried for disposal with the technical standards [13].

If a sample is, during the inspection, collected by destroying a waste form that has already been fabricated or obtained from a waste form in order to verify the compliance with the technical standards, sampling time and costs will be required; in addition, the quality of the waste form may be degraded. For these reasons, a verification method that utilizes process data from the fabrication procedure of waste forms and other relevant data is generally used in order to prove the compliance with the technical standards of the waste forms. In other words, this is based on the notion that the quality of the waste forms is secured by using various types of data and information in a comprehensive manner. Specifically, these data and information include data and waste information from non-destructive measurements that can be performed relatively easily; quality control data concerning the waste form fabrication such as operating records of solidification facilities and test reports on the used solidification materials; and the data which are collected in advance to supplement the previously mentioned information and data. The data collected in advance correspond to waste form performance assessment data, sampling analysis data (nuclide-specific radioactive characteristics, properties of solidified forms), and the correlation coefficients. Accordingly, the waste form verification with this method requires quality control and data acquisition in the course of the waste treatment and solidification process.

Finally, as of today, there are no specific technical standards for high-level radioactive waste forms. The only stipulation requires that these materials be solidified or confined in containers.

Exercises

1. When managing radioactive wastes, interdependencies of individual management processes need to be kept in mind (Principle 8 of the IAEA *Principles of Radioactive Waste Management Safety Fundamentals*). Consider an example in which radioactive liquid waste that is relatively high in radioactivity concentration and contains sulfate undergoes an evaporation and is subsequently cemented. List and describe issues for which interdependency may be a concern and provide appropriate solutions for these issues.
2. Consider the following case: high efficiency particulate air (HEPA) filters are placed in two layers for treating off-gas that contains 0.6 $\mathrm{Bq/cm^3}$ of ^{60}Co. The off-gas is released to the environment after attaining a level that is 1/100 of the emission standard (3×10^{-6} $\mathrm{Bq/cm^3}$). Calculate the collection efficiency required for the HEPA filters. Assume that filters of the same collection efficiency are used for the two layers and all of the ^{60}Co in the off-gas is present in particulate form.
3. Suppose that flammable cellulosic solid waste contaminated by ^{3}H (tritium) and ^{60}Co are treated by incineration, and there is an intention to reduce the amount of radioactive material released to the environment upon emitting gaseous waste generated from the incineration. In this situation, which treatment method should be selected, and what is the basis of this selection? In addition, name any secondary wastes that may be generated in the course of gaseous waste treatment and identify any treatment methods that are considered appropriate for treating the secondary wastes. Finally, if the flammable waste is vinyl chloride and undergoes incineration treatment, what precautions are required?
4. Describe the strengths and weaknesses of compaction and melting as applied to non-flammable solid wastes from the viewpoint of burial disposal.
5. Describe the difference between the gaseous wastes generated at nuclear power plants and those generated at reprocessing plants. Also describe the difference in the treatment methods of these gaseous wastes due to their differences in characteristics.
6. Describe the reasons why vitrification is used as a technique to solidify high-level radioactive liquid wastes.

References

1. International Atomic Energy Agency, *Principles of Radioactive Waste Management Safety Fundamentals*, Safety Series No. 111-F (1995)
2. Radioactive Waste Management Center, *Changes in Generation and Treatment of Waste from Nuclear Power Reactor Operations*, RWMC Topics, No. 22 (1992) (in Japanese)

3. W.E. Browing, C.C. Bolta, *Measurement and analysis of the holdup of gas mixtures by charcoal adsorption traps*, ORNL-2116, Oakridge National Laboratory (1956)
4. A. Suzuki, R. Kiyose, *Nuclear Fuel Cycle Engineering* (Nikkan Kogyo Shimbun, Tokyo, 1981), p. 113 (in Japanese)
5. International Atomic Energy Agency, *Application of Membrane Technologies for Liquid Radioactive Waste Processing*. IAEA Technical Reports Series No. 431 (International Atomic Energy Agency, Vienna, 2004)
6. International Atomic Energy Agency, *Handling and Processing of Radioactive Waste from Nuclear Applications*. Technical Reports Series No. 402 (International Atomic Energy Agency, Vienna, 2001)
7. International Atomic Energy Agency, *Application of Ion Exchange Processes for the Treatment of Radioactive Waste and Management of Spent Ion Exchanges*. IAEA Technical Reports Series No. 408 (International Atomic Energy Agency, Vienna, 2002)
8. International Atomic Energy Agency, *Application of Thermal Technologies for Processing of Radioactive Waste*. IAEATECDOC-1527 (International Atomic Energy Agency, Vienna, 2006)
9. A. Chrubasik Dipllng, Development of an Incineration System for Radioactive Waste, in *Radioactive Waste Management 2* (British Nuclear Energy Society, London, 1989)
10. M. Suyari, R. Nakanishi, T. Noura, M. Fujitomi, S. Ano, An incineration technology for low level radioactive solid waste. R&D Kobe Steel Eng. Rep. **53**(3), 56–60 (2003) (in Japanese)
11. N. Kuchiki, S. Fukuda, T. Kakuta, K. Sato, S. Yamazaki, Development of high-frequency induction melting system for radioactive waste disposal. FAPIG **175**(7), 18–22 (2007) (in Japanese)
12. Radioactive Waste Management Center, *Current State of Treatment, Storage and Disposal Technologies*, RWMC Topics, No. 25 (1993) (in Japanese)
13. T. Yagi, N. Takebayashi, Outgoing inspection technology for low-level radioactive waste. Nucl. Eng. **35**(4), 60–64 (1989)

Chapter 6
Radioactive Waste Disposal

Yuichi Niibori

6.1 Concept of Radioactive Waste Disposal

The first concept of geological disposal proposed in history is probably the direct disposal of high-level radioactive liquid wastes in salt formations indicated in a 1957 report [1] prepared by the U.S. National Academy of Sciences (NAS). The concept, however, differs considerably from today's concept of geological disposal in that, for example, the plan in those days was to directly inject liquid waste and the time span considered was only 600 years. The basic ideas of today's disposal systems are from the concept indicated in the so-called Polvani Report [2] in the 1970s and the KBS concept [3] developed in Sweden in the early 1980s. The basis, therefore, had been established by the end of the 1980s. Today, R&D on geological disposal systems is underway in more than 30 countries [4].

R&D in the past 30 years has encompassed diverse approaches. If geological disposal were thought of merely as a very large scale construction project involving excavating an underground tunnel, putting waste into corrosion-resistant containers and emplacing the waste containers, it would have been possible to carry out geological disposal quickly. With the progress of research and development and the deepening of social debate, however, it has become a common understanding that the greatest concerns relate to concepts; how safety can be maintained over a

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The nuclear regulatory system in Japan has been changed significantly after the Fukushima Daiichi Nuclear Power Station accident in March 2011. Descriptions in this chapter have been translated from the book originally published in Japanese before the accident, with minimal update where appropriate. The Nuclear Regulation Authority newly established after the accident has not completed its review for the guidelines and regulations established by the former Nuclear Safety Commission. In this chapter, guidelines set by the NSC have been adopted.

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long period of the order of longer than 10,000 years, how long-term system performance can be verified and regulated and how such efforts can be incorporated into the framework for social consensus building. The process that guides such R&D and social debate is performance assessment, which is explained in Chap. 7, “Performance Assessment of the Radioactive Waste Disposal System.”

6.1.1 Basic Concepts for Ensuring Safety

The type of disposal of radioactive wastes varies depending on the waste types, but in any case, the type of disposal must be such that the exposure of the general public can be kept at or below the specified limits and can be made as low as reasonably possible. This means that an approach from the viewpoint of radiation protection is important. By regarding this as a basic safety requirement, in order to ensure the safety thus defined, a radioactive waste repository usually forms a “multi-barrier structure” consisting of “engineered barriers” including waste forms themselves and “natural barriers” including surrounding formations. Natural barriers mainly have the function of maintaining the stability of engineered barriers over a long period of time or enabling engineered barriers to function normally. Engineered barriers, which are designed in view of the function of natural barriers, have the function of retaining radionuclides as long as possible and slowing down the release of radionuclides to natural barriers. Thus, these barriers are expected to be mutually complementary. When we use the term “disposal system,” we are referring to the entirety consisting of waste forms and the multiple barriers surrounding them.

Every organization and institution disposing of radioactive wastes must evaluate the potential impact of the wastes in the planned disposal system on the general public and show that it will not pose any problem in ensuring safety. The assessment of the impact on the general public begins by classifying possible future events into a number of scenarios and identifying the conditions constituting those scenarios (see Sect. 7.1.2). Since the disposal system is supposed to stay in a sound condition over a long period of time and therefore needs to be evaluated with respect to performance after the active management period, it must be kept in mind that there is some degree of inherent uncertainty in the anticipated events. It is common practice to assume scenarios involving natural processes and those involving artificial processes. These scenarios are classified into highly probable, realistic scenarios and low-frequency or even rare scenarios, and the disposal system is evaluated from the viewpoint of radiation hazard prevention. Although scenarios in the latter category may not be realistic, they are used to judge whether the design of the repository under consideration is capable of coping with a wide range of uncertainties. This kind of approach, which takes into consideration the possibility of occurrence of various scenarios, is called a “risk-based approach.” Specific examples are given in Sect. 6.3. The risk-based approach [5] that evaluates the impact of a scenario by taking into account the possibility of occurrence of that scenario has been adopted in many countries, and efforts are underway to establish systems based on that approach.

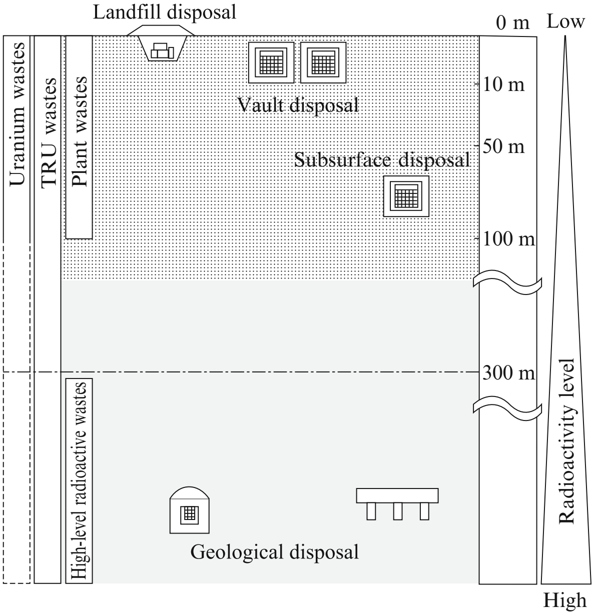
One of the most realistic scenarios involving natural processes is a groundwater scenario. This scenario, which assumes that radionuclides migrate out of the repository as part of groundwater flow and reach the biosphere, is one of the most important scenarios in safety assessment. Other scenarios attributable to natural processes other than the groundwater scenario include scenarios describing the possibility of radionuclides reaching the biosphere because of earthquakes, volcanic activities or slower processes such as upheaval and erosion. These scenarios also affect the migration of radionuclides due to groundwater. Scenarios involving artificial processes assume the occurrence of radiological hazards due to radioactive wastes buried at a repository, which results when, for example, such wastes are disturbed unintentionally because records are no longer available about the existence of the repository; for example, excavation activities or land reuse activities carried out for the purpose of mineral or energy resources (including water resources) development or surveys. These are called “human activity scenarios.” Although it is not possible to predict human activities in the distant future, standard human activities are assumed (this is called “stylization” and presented in Chap. 7) on the basis of other information such as current knowledge, and the soundness of the disposal system under consideration is checked from the viewpoint of radiation hazard prevention. It is also necessary to study radiation protection during repository operation. Scenarios in this category are classified as “operation scenarios” and are distinguished from the scenarios mentioned above.

All scenarios require a thorough discussion involving stakeholders as central participants and transparency with respect to the framework for and the scope and significance of such discussion. In view of the purpose of protecting the health of the general public from nuclear projects, it is an international consensus that the scenario in which someone intentionally destroys a repository and becomes exposed to radiation is not taken into consideration in assessing the safety of repository projects.

6.1.2 Disposal Methods for Different Types of Wastes

As mentioned in Sect. 1.1.2, radioactive wastes vary widely. In Japan, basically, radioactive wastes are disposed of at Category 1 waste disposal facilities or Category 2 waste disposal facilities depending on the level of radioactivity involved. Here “Category 1” and “Category 2” are not classifications of waste; instead, they refer to different burial concepts. Category 1 waste disposal facilities, which are designed for so-called geological disposal, are used in cases where wastes need to be isolated from the biosphere over a long period of time. In this disposal method, which is described in detail later in this chapter, wastes are disposed of at depths greater than 300 m from the ground surface. The types of wastes to be disposed of by this method are high-level radioactive wastes and some low-level radioactive wastes known as TRU wastes. Category 2 waste disposal facilities, which are designed for management type disposal, are used for relatively shallow depth burial of low-level radioactive wastes other than the types mentioned above. Different methods are illustrated in Fig. 6.1 of which there are four types of

Fig. 6.1 Four types of disposal methods in Japan (http://www.enecho.meti.go.jp/rw/gaiyo/gaiyo03.html)



disposal: landfill disposal, vault disposal, subsurface disposal, and geological disposal. Landfill disposal is burial disposal that does not use any artificial structure, while vault disposal is burial disposal that uses a concrete vault placed at a relatively shallow depth. Subsurface disposal is disposal at depths sufficiently greater than the depths at which ordinary underground space use takes place (greater than 50 m). These three types of disposal correspond to Category 2 waste disposal facilities. High-level radioactive wastes are liquid wastes generated after uranium and plutonium extraction at a reprocessing plant and they are solidified with borosilicate glass. This includes the high-level radioactive wastes generated by reprocessing that has been commissioned by Japan to France and the U.K. Reprocessing also generates non-high-level radioactive wastes such as hulls, end pieces, waste silver absorbent and concentrated liquid wastes. The same thing occurs even when reprocessing is commissioned to overseas contractors because the wastes generated by contracted reprocessing are also returned. An option currently under study is to return vitrified high-level radioactive waste that has a similar level of radioactivity but has a much smaller volume, instead of returning the aforementioned voluminous wastes.

6.2 Multi-barrier Approach

The waste disposal system consists of not only waste forms but also artificially installed barriers and the surrounding geological environment. The former are called engineered barriers, and the latter, natural barriers. Engineered barriers vary depending on the wastes to be disposed of. Natural barriers are expected not only to physically isolate radionuclides from the biosphere but also to provide stable external conditions needed to keep the engineered barrier environment unchanged over a long period of time. Conditions to be kept unchanged include hydraulic conditions such as groundwater flow to engineered barriers, chemical conditions such as dissolved oxygen content of groundwater, and mechanical conditions such as stresses acting on tunnels, backfill material and canisters.

A quick review of the results of performance assessments made in other countries confirms that many of the radionuclides contained in the disposed of wastes decay sufficiently in the engineered barriers or in relatively limited regions around the barriers. The role of engineered barriers is to restrict the flow of groundwater into the waste forms and retard the migration of radionuclides. External conditions for engineered barriers are determined by natural barriers, and those conditions change over time. Within engineered barriers also, changes are likely to occur over a long period of time. When designing an engineered barrier, it is necessary to take adjustment with natural barriers into consideration. On the natural barrier side of the engineered/natural barrier boundary, there is a region where disturbance occurs because of the installation of the engineered barrier. This region is called the excavation-damaged zone. Factors contributing to disturbance may include excavation-induced cracking in rock, resultant changes in groundwater flow and stress distribution, and changes in chemical conditions resulting from those changes. The region consisting of the engineered barrier and the excavation-damaged zone, therefore, is sometimes called the “near field,” and the natural barrier that is not affected by the installation of the engineered barrier is called the “far field.”

There are countries that have rules requiring the adoption of the best practical means (BPMs) in the design, construction and verification of repositories or the implementation of the best available techniques (BATs) besides the approaches described above. An example of the former countries is the U.K., and of the latter, Sweden. Repositories differ from ground-level facilities in that it is difficult to directly observe repositories. The goal of these approaches, therefore, is to make the risks to future generations of the general public as small as reasonably possible by using the best possible means. The risk-based approach mentioned in the preceding section, which combines the probability of occurrence of particular scenarios and their hazards, aims to cope with the uncertainty inherent in the disposal system. It can be said that what the multi-barrier approach aims to achieve is to work out a comprehensive solution by considering such ideas as BAT together with the mutual complementation of different types of barriers.

In the case of a nuclear reactor, too, multiple barriers are installed. In the case of a multi-barrier system for a reactor, it is assumed that the stage (barrier) preceding each barrier can be completely destroyed. This concept is called “defense-in-depth.” In the case of a multi-barrier system for geological disposal, if a barrier is broken and radionuclides are released, it is not thought that the barrier function has been completely lost. Instead, it is thought that the barrier system remains functional and effective in isolating the radionuclides from the biosphere. For example, even if the overpack has been corroded to the extent that a hole is formed so that the waste forms inside and the infiltrating groundwater come into contact and the radionuclides have begun to leak out of the canister, the canister and its corrosion products are in many cases effective in keeping the groundwater in the engineered barrier chemically reducing. Thus, each barrier is not considered independently. The defense-in-depth concept for geological disposal differs from that for reactors in that the former takes into consideration mutual complementation among barriers. Multi-barrier approach can achieve such mutual complementation because each of the barriers used for geological disposal not only has the function of physically containing radionuclides but also performs chemical and mechanical functions.

6.3 Near-Surface Disposal: Category 2 Waste Disposal

In this chapter, the term "near-surface disposal" refers to Category 2 waste disposal classified as landfill disposal, vault disposal or subsurface disposal. The use of scenarios to ensure safety has been mentioned earlier. In Japan, a realistic groundwater scenario is used as a basic scenario, and disposal system assessment is made under that realistic scenario from the viewpoint of radiation protection. In the case of low-level radioactive wastes to be disposed of at shallow depths, dose levels for the general public at which a disposal system can be deemed in a sound condition under the basic scenario are 10 μSv/y or less. Doses of 10 μSv/y or less are sufficiently low so that management is not necessary from the viewpoint of exposure management. This is one of the lowest target values in the international community [5]. When the design of a disposal system is evaluated to check if it is capable of coping with a wide range of uncertainties, a rough criterion of 300 μSv/y or less is used.

In Japan, staged management is applied to these disposal methods. The term "staged management" refers to an active management approach in which, in order to make exposure doses for the general public as low as reasonably possible, the waste burial site is managed in stages according to waste characteristics such as the type of radioactive solid waste and radioactivity level until it is confirmed that the influence of the radioactive material on the accessible environment from the viewpoint of safety has become sufficiently low mainly because the radioactivity level of the buried radioactive solid wastes has decreased over time. In staged management, basic safety functions needed to ensure the safety of the burial facility concerned are determined according to the disposal method to be used. The basic safety functions can be classified [6] as follows.

1. Dispersal prevention: This function is needed to prevent radioactive materials from being dispersed out of the burial site for wastes such as waste concrete.
2. Shielding: This function is needed to cut off radiation and reduce the influence of radiation on the surrounding area.

3. Closure: This function is needed to prevent radioactive materials from leaking from artificial structures of waste burial facility.
4. Suppression of migration: This function is needed to reduce the exposure of local residents by suppressing the groundwater-mediated migration of radionuclides leaking from a waste burial site to the living environment.
5. Isolation: This function is needed to prevent or reduce exposure due to accidental entry of humans by burying radioactive wastes at a depth (distance) sufficiently greater than the depths at which ordinary underground space use takes place.

Basic safety function requirements vary depending on the type of disposal. The length of the period during which necessary functions are to be maintained, the facility designs to be taken to do so, and the monitoring method are determined accordingly.

6.3.1 Landfill Disposal

In landfill disposal, waste forms of very low-level radioactive wastes—such as concrete and metals—that are chemically and physically stable are placed in an unlined near-surface trench. Management is performed in two stages: the burial stage, from waste form emplacement to the completion of covering with soil; and the maintenance stage, from the completion of covering with soil to the completion of the management period. After a required period of time, the land becomes available for ordinary uses. This type of disposal has been carried out on an experimental basis on the premises of the Japan Atomic Energy Agency (JAEA) for the waste generated by the decommissioning of the Japan Power Demonstration Reactor. Figure 6.2 shows a general view of the waste burial experiment site. The Morvilliers disposal facility in France, which went into operation in 2003, is a commercial example in this category.

Waste disposed by this method is characterized by very low-level radioactivity and a large quantity. What is important, therefore, is how the volume of wastes can be made as small as possible before burial at the disposal site. The quantity of wastes generated by the decommissioning of a commercial power reactor is estimated to be 3,000–10,000 tons per reactor though it varies depending on the type of reactor and the magnitude of power generation capacity. The decommissioning of a nuclear power plant generates about 500,000–550,000 tons of waste concrete and metals, and very low-level radioactive wastes account for about 2 % of that quantity (see Sect. 3.1.2).

The safety-related functions required in landfill disposal are shielding and the suppression of migration at the burial stage and the suppression of migration at the next (maintenance) stage. Dispersal prevention is also required at the time of waste form emplacement at the burial stage. Whether or not the shielding function

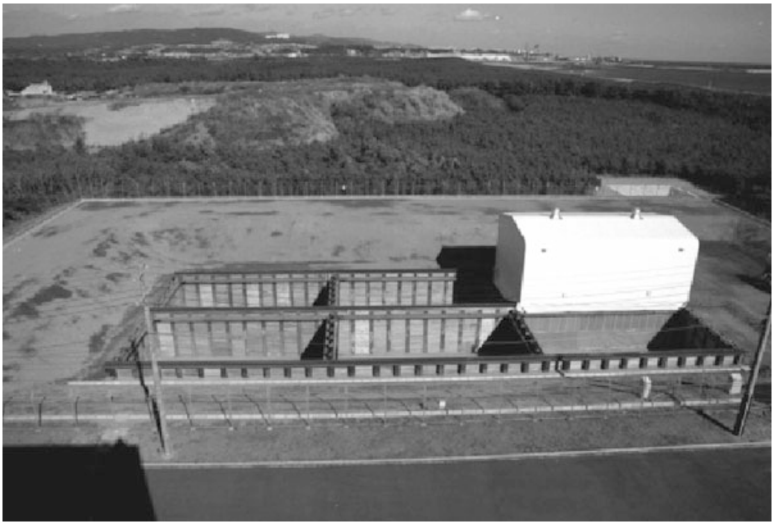


Fig. 6.2 General view of JAEA's waste burial field test facility (http://www.enecho.meti.go.jp/rw/gaiyo/gaiyo03-1.html)

is effective is evaluated by measuring the dose equivalent in the environmental monitoring area and checking if the dose limit is exceeded or not. The migration-suppressing function at the same stage aims to prevent the occurrence of voids (that can cause large deformations such as cave-ins) and the exposure of buried items. In order to achieve these goals, control items are defined for the execution of covering with soil to monitor the covering work, and the concentration of radioactive materials in groundwater is measured in the environmental monitoring area to check if migration to the living environment is being suppressed successfully. If it is judged that these basic safety functions are not working as expected, remedial actions such as repair need to be taken. At the maintenance stage, control items of the migration-suppressing function will include defining, patrolling and inspecting the burial conservation area and installing and maintaining signage, etc. for the purpose of banning or restricting particular acts such as agricultural activities. The management process proceeds from the burial stage to the maintenance stage, when covering with soil has been completed and the stability of the soil covering has been confirmed.

6.3.2 Vault Disposal

Vault disposal is a method of burying radioactive wastes in an artificial structure such as a concrete vault constructed at a relatively shallow depth in the ground. Wastes for which this disposal method can be used include concentrated liquid wastes, spent ion exchange resin and incineration ash generated by burning combustible items solidified with cement and packed in drums, and solid wastes such as pipes and filters. These are relatively low-level radioactive wastes mainly from nuclear power plants. In this disposal method, the management process is divided into three stages according to radionuclide concentrations and the progress of work. The first stage is from the emplacement of waste form to the completion of covering with soil. The period following the completion of covering with soil until the completion of the management period is divided into the second and third stages according to the categories of control measures. In Japan, these stages are taking place at the Low-Level Radioactive Waste Disposal Center of Japan Nuclear Fuel Ltd. (JNFL) located at Rokkasho Village in Aomori Prefecture. It is thought that the management period continues for 300–400 years. The JNFL Rokkasho facility was designed by referring to the low-level radioactive waste disposal facility in Aube, France. It is designed that rainwater flowing through the facility is collected from the inspection tunnels to monitor any contamination.

The basic safety functions required in vault disposal are shielding and closure at the first stage and the suppression of migration at and after the second stage. At the first stage, control measures related to the soil-covering work are also taken so that migration can be suppressed effectively at and after the second stage. The closure function checks if there is significant leakage by measuring the concentration of radionuclides in the surrounding groundwater. At the second stage, the concentration of radionuclides in the surrounding groundwater is measured and monitored to check if migration is being suppressed effectively. Measures taken at the third stage are the same as at the maintenance stage of landfill disposal, i.e., defining, patrolling and inspecting the burial conservation area and installing and maintaining signage, etc. for the purpose of, for example, banning or restricting particular acts such as agricultural activities.

Figure 6.3 illustrates the concept of the Low-Level Radioactive Waste Disposal Center of JNFL. The waste form is cemented waste packed in a drum, and the space

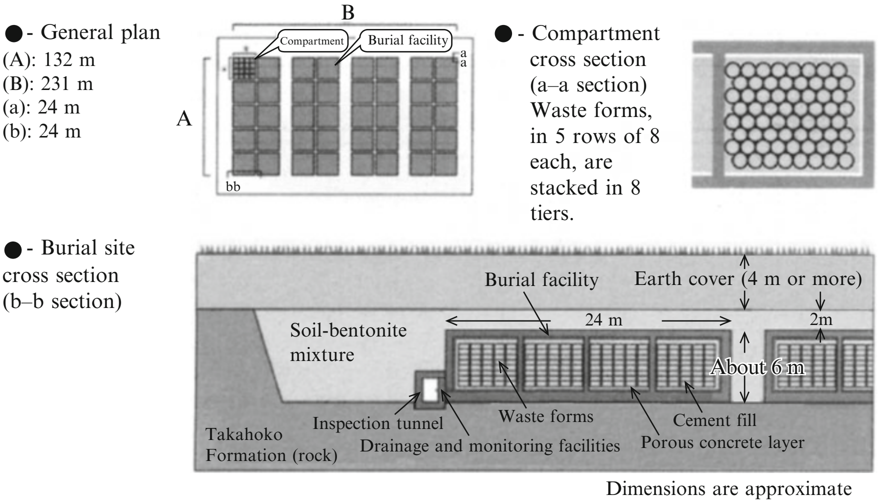


Fig. 6.3 Concept of vault disposal (burial facility #1 at JNFL) (http://www.rjnfl.co.jp/business-cycle/2_maisetsu/maisetsu_03/maisetsu_04/maisetsu_04_04.html)

in the vault (the “Burial Facility” in the figure) is also filled with cement. The space around the vault is filled with mixed soil (a mixture of about 15 % bentonite and excavated soil) to make the permeability of the mixed soil zone lower than that of the surrounding rock. Then, the top of the vault is covered with earth and soil, and these covering materials are compacted. Bentonite is a type of rock composed mainly of a mineral called montmorillonite and also containing other minerals such as quartz, mica, feldspar and zeolite. The ion exchange property of montmorillonite enables to retain cations. By absorbing water, bentonite increases its volume by more than ten times, and its low permeability can be used to form a low-permeability groundwater barrier. The bentonite–soil mixture is an engineered barrier material that is not used at the Aube facility. The Aube disposal facility has monitoring tunnels to periodically check on rainwater infiltration.

JNFL began its waste burial operation in 1992. As of the end of July, 2010, about 221,000 drums have been buried. The current plan is to bury 1,000,000 drums, but a capacity increase to about 3,000,000 drums is currently under study.

6.3.3 Subsurface Disposal

“Subsurface disposal” in the Japanese disposal categorization is a method of disposal in which a tunnel-type or silo-type underground concrete structure is constructed at a depth (50–100 m below ground surface) sufficiently deeper than the depths at which ordinary land uses (e.g., habitation) and underground space utilization (e.g., the construction of a basement to support an aboveground structure, use of underground space for subways, drinking water and sewerage systems, utility tunnels and building basements) take place, and waste forms are buried in that structure. Wastes to be disposed of by this method include reactor core internals such as shrouds, channel boxes and spent control rods [7]. A shroud is a cylindrical structure to house fuel assemblies and control rods constituting the reactor core. Using jet pumps, the shroud serves as a partition to help form coolant flow channels toward the reactor core from under the shroud. A channel box is a square metal tube housing fuel assemblies. These wastes are classified as low-level radioactive wastes with relatively high radioactivity concentrations.

Subsurface disposal is divided into the construction and burial stage and the post-backfilling stage. The basic safety functions required at the former stage are shielding and closure, while the function required at the latter stage is the suppression of migration. In subsurface disposal, isolation, which is one of the basic safety functions, is required continuously even after the end of the management period. When selecting control items in each case, therefore, it is necessary to select them so that appropriate functions are available when needed. In subsurface disposal, safety reviews are made by the operator at intervals not longer than 20 years during the staged management process. The purpose of these reviews is to check whether the functions of the entire disposal system including the natural barrier meet the functional requirements even if the individual barrier functions have deteriorated

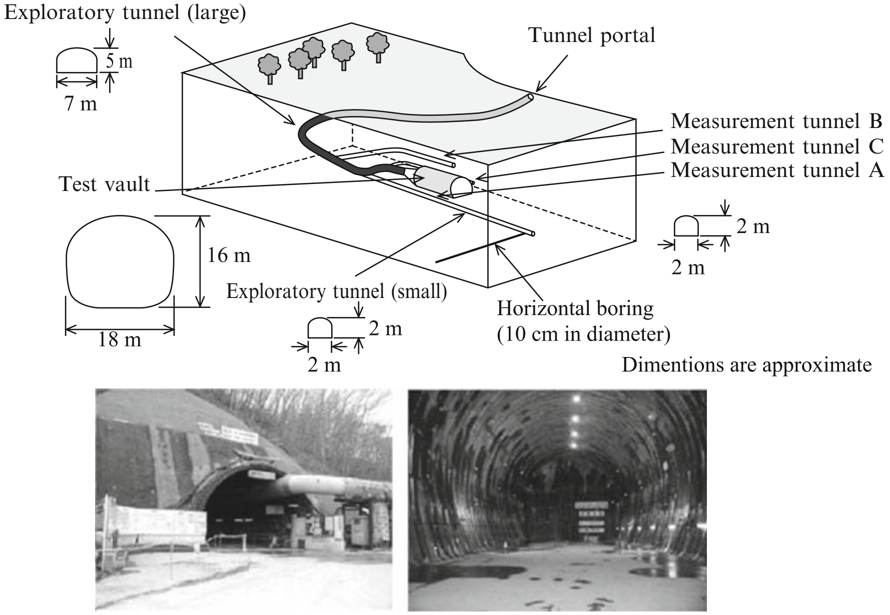


Fig. 6.4 Subsurface disposal test silo of JNFL: overview (*top*), tunnel portal (*bottom left*) and test vault (*bottom right*) (http://www.enecho.meti.go.jp/rw/gaiyo/gaiyo03-3.html)

over time. In the event of a beyond-design-basis behavior of the entire system, it is, of course, necessary to make appropriate repairs.

The preliminary study conducted from July 2001 to June 2002 on the premises of JNFL's Low-Level Radioactive Waste Disposal Center indicated that the construction of a test silo was feasible, and a detailed study (November 2002 to March 2006) was conducted to obtain comprehensive geological and groundwater-related information needed to design a burial facility. Figure 6.4 illustrates the test silo for subsurface disposal. As shown in the illustration of the upper half of the figure, there is a tunnel leading from the tunnel portal (bottom left photo) to the test silo. This is called an access tunnel. By using a vault (silo) as shown in the bottom right photo, waste forms are emplaced in a concrete vault. Figure 6.5 shows the concept of engineered barriers for subsurface disposal. In the case shown here, concrete is placed at the bottom, and a vault is completely enclosed by a low-permeability layer consisting of bentonite blocks and then by a low-diffusion layer made of cement-based material is installed. Then, as waste forms are placed in the vault, the void space in each compartment is filled with cement. After a predetermined number of waste forms have been emplaced, the vault is closed with a lid, and on that lid, a low-diffusion layer lid and then a low-permeability layer lid are placed. The void space along the sides is backfilled with cement, and the void space over the waste forms is backfilled with fill material prepared by using, for example, excavated material. Although the purpose of the low-permeability layer (bentonite layer) is to reduce groundwater inflow, it also contributes considerably to the reduction of diffusion in the migration of radionuclides [8].

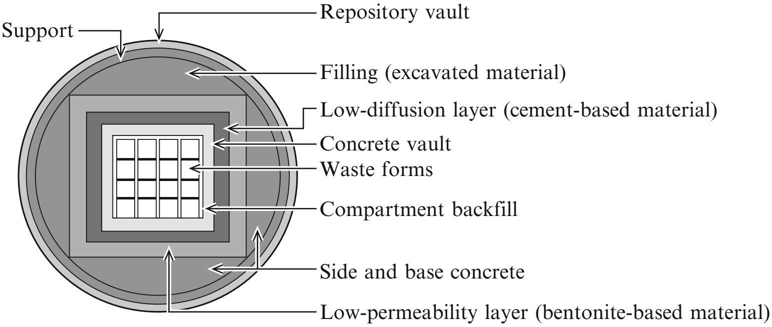


Fig. 6.5 Engineered barriers for subsurface disposal [8]

It is generally said that the period of safety assessment for subsurface disposal is 10,000 years or longer [8, 9]. The classification of scenarios including groundwater movement, upheaval, erosion and low-frequency events as well as reviews of study results have been underway, and the obtained results provide feedback for the construction of engineered barriers and the emplacement of waste forms.

6.4 Geological Disposal: Category 1 Waste Disposal

Geological disposal differs considerably in concept from the management-type disposal approaches, in that geological disposal aims to ensure long-term safety without relying on human involvement. In other words, it is a passive approach [10]. Needless to say, as in subsurface disposal, safety reviews are conducted periodically during the period from the emplacement of waste forms through engineered barrier construction, backfilling and closure to decommissioning. When disposing of the waste types to which geological disposal is applied, it is necessary to keep the influence on the accessible environment of the radiation due to the radionuclides concerned at or below the required level over a long period of time beyond the limit that can be placed under human management. That period is said to be longer than several hundred thousand years. In order to manage something over such a long period of time, it is necessary to use a passive method of disposal that does not rely on human involvement. The Nuclear Energy Agency of the Organisation for Economic Co-operation and Development (OECD/NEA) [11] stresses the importance of not only protecting human health and the environment but also minimizing the burden on future generations by using geological disposal systems. With respect to burdens on future generations, basically, the cost of disposal should be borne by current beneficiaries. The intergenerational balance should be determined according to the level of risk that may be left to future generations. This means that the management and disposal of things that need to be isolated from the biosphere for a long period of time such as high-level radioactive wastes should not be left to future generations. Instead, basically such management and disposal are the current generation's responsibility and should be funded by the current generation. In order to cope with risks that are nevertheless left to future generations, it is necessary to build disposal systems so that the level of risk to be transferred is acceptable to future generations.

The industrialization of the geological disposal of high-level radioactive wastes in Japan is based on the Specified Radioactive Waste Final Disposal Act, which was enacted in June 2000. The law aims to create an environment for nuclear power generation by taking measures necessary for the final disposal of specified radioactive wastes (high-level radioactive wastes, or vitrified wastes) generated by the reprocessing of spent fuel, in order to contribute to proper use of nuclear energy for power generation. The basis for this law dates back to the policy indicated by the Japan Atomic Energy Commission in 1976 [12].

The process that has led to the present situation, including notable events in Japan and abroad, is summarized below. The Specified Radioactive Waste Final Disposal Act was amended in June 2007. The amended law classified different disposal methods into Category 1 waste disposal and Category 2 waste disposal. Under the amended law, the organization that carries out disposal can handle not only vitrified wastes attributable to domestic spent fuel but also foreign vitrified wastes received in exchange for low-level radioactive wastes, and TRU wastes that need geological disposal.

The 1976 policy of the Japan Atomic Energy Commission called for attaching importance for the moment to geological disposal and conducting research on disposal methods suitable for particular social and geographic conditions. This policy was adopted in view of the growing interest in environmental issues in each country and the ongoing discussions on the disposal of spent fuel. In 1977, OECD/NEA proposed "Objectives, Concepts and Strategies for the Management of Radioactive Waste Arising from Nuclear Power Programmes," [2] which is still one of the conceptual bases for geological disposal. The document made the following key points.

1. Any type of geological environment can be regarded as a possible host for a repository as long it satisfies the basic requirements.
2. The safety of geological disposal should be ensured not only by the natural environments, but also by a multi-barrier system which includes engineered components of the repository.
3. Rational, scientific and realistic evaluation is essential for ensuring the long-term safety of disposal.

With these points in mind, an international cooperative research project was conducted from 1980 to 1992 at the site of the Stripa mine in Sweden. Concurrent with this and other laboratory-level research efforts to gain basic knowledge, efforts have also been made to enhance the level of technology [10].

In 1987, the Swedish National Council for Nuclear Waste (KASAM) and the National Board for Spent Nuclear Fuel (SKN) conducted a study on ethical aspects of problems associated with uncertainty in geological disposal [13, 14]. Focusing on problems related to the responsibility of current generations for future generations and the right of future generations to make their own decisions, the study concluded that the responsibility of current generations should not be passed on to future generations, but the possibility of involvement of future generations should not be denied. In the 1990s, reviews were made of the slower-than-expected progress of geological disposal plans in some countries. For example, in 1990, the National Research Council (NRC) [15] of the U.S. National Academy of Sciences stated that because geological disposal systems, unlike other engineering systems, need to work with natural environments, projects need to be carried out in stages in view of site investigation results, and that the evaluation of the safety of geological disposal systems should be distinguished from the prediction of future behavior. This was a warning about the fact that some researchers were discussing safety assessment and behavior prediction in a confusing manner so as to unnecessarily complicate discussions on safety. The NRC also stated that the safety of repositories was in some cases not only a scientific, technical issue but also an issue to be judged against social criteria.

Thus, the safety of geological disposal needs to be evaluated from various viewpoints. The entirety of arguments and evidence for safety is sometimes called a "safety case." According to the definition given by OECD/NEA [16], a safety case is the synthesis of evidence, analyses and arguments that quantify and substantiate a claim that the repository will be safe after closure and beyond the time when active control of the facility can be relied on. According to IAEA's definition [17], a safety case also includes the safety of workers during operation.

Basically, the content of what is called a safety case is to be built according to the situation of each country, but it must hold in international discussions. It is an integration of knowledge from a variety of disciplines including nuclear engineering, earth sciences, civil engineering, chemistry, chemical engineering and social sciences. To be more specific, it includes not only knowledge related to the characteristics of engineered barriers and natural barriers and their uncertainty needed for dose assessment but also studies on regions affected by volcanoes and active faults as well as attempts to indirectly verify the long-term performance of disposal systems through natural analogue studies. The term "natural analogue study" means a study which aims to learn, for example, about the preservation and migration processes of radionuclides by regarding uranium ore deposits as something analogous to geological disposal facilities or to investigate the alteration processes of barrier materials by studying natural phenomena. The research on the natural reactors at Oklo [18, 19] is a famous example. It has been found that at the uranium ore deposits at Oklo in the Haut-Ogooué province of Gabon, some 2 billion years ago, spontaneous fission (a reaction corresponding to an average output of 100 kW) occurred over a period of several hundred thousand years, and by studying the site, it is possible to learn about the natural preservation and migration processes of radionuclides. There is also a need for a wide range of viewpoints such as integrating experimental data (developing a database system) on the migration of radionuclides; developing methodologies for enduring the transparency of such data; identifying differences in the disposal facility layout between the case in which pre-closure retrievability of waste forms and reversibility of operation are taken into consideration and the case in which they are not taken into consideration; and taking account of the future of nuclear fuel cycles, e.g., the disposal of high-level radioactive wastes from both fast breeder reactors and light water reactors during the period of combined use of those types of reactors (see for example, Refs. [20, 21]).

The types of radioactive wastes for geological disposal in Japan are, as mentioned earlier, vitrified high-level radioactive wastes produced by vitrifying high-level radioactive liquid wastes from the reprocessing of spent fuel, and some of TRU wastes classified as low-level radioactive wastes. Wastes in the latter category, TRU wastes, because they are characterized by the presence of transuranium elements which are heavier than uranium, such as neptunium, plutonium, americium and curium. The long-lived low-decay-heat-generating radioactive wastes that need geological disposal include spent fuel hulls from reprocessing plants, fuel assembly end pieces, silver adsorbent that has adsorbed iodine, and wastes generated by the decommissioning of reprocessing plants or MOX fuel fabrication plants. These wastes contain long-lived nuclides of iodine (^{129}I) and carbon (^{14}C) as well as transuranium elements [22].

The base material of vitrified high-level radioactive wastes is borosilicate glass. Vitrified waste forms are made by putting neutralized high-level radioactive liquid wastes mixed with molten borosilicate glass into stainless steel canisters at a reprocessing plant. Vitrified waste forms are black in color and opaque. As an example, vitrified waste forms will be cooled at the High-Level Radioactive Waste Storage Facility of JNFL at Rokkasho Village, Aomori Prefecture for 30–50 years, and then will be buried at the underground repository selected for disposal.

If about 30 tons of spent fuel generated from one year of operation of a 1,000 MWe nuclear power plant is reprocessed, about 0.9 tons of high-level radioactive liquid waste is generated. This translates to about 30 vitrified waste forms. If it is assumed that nuclear power generation accounts for about 1/3 of Japan's electricity generation and the burnup of fuel is 45,000 MWd/MTU, the volume of high-level radioactive wastes generated from the electricity consumed by one person in Japan during his or her lifetime (80 years) is about 0.1 liter (see Exercise 2 at the end of this chapter).

In Japan, geological disposal facilities are sited at places free from volcanoes, active faults and useful underground resources. At depths of 300 m or greater under such locations, engineered barriers are installed, and waste forms are disposed of. Needless to say, even at depths of 300 m or greater, if an aquifer including remarkably fast flow of groundwater is found, then a disposal facility is constructed in an even greater depth.

Figure 6.6 shows the concept of geological disposal. In geological disposal, the canister mentioned earlier is put into an overpack, which is then enclosed in bentonite. Because the overpack is surrounded by a natural barrier, this is a typical example of the multi-barrier system mentioned in Sect. 6.2. As a design option, it has been proposed that 40,000 vitrified waste forms be emplaced in an underground

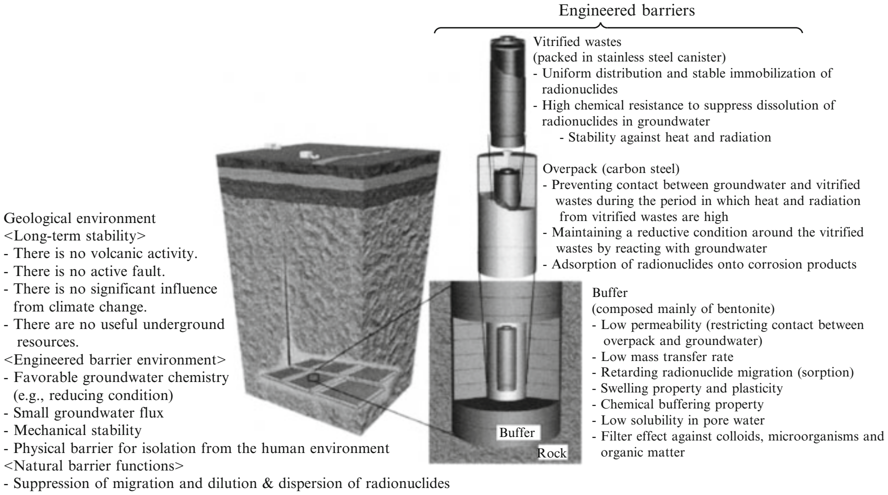


Fig. 6.6 Geological disposal system (functions of engineered and natural barriers) [23]

repository area measuring about 2 km by 3 km. The repository area is determined taking into consideration a number of factors such as suppressing the alteration of engineered barriers and the boiling of groundwater; dimensional requirements for engineered barriers; restrictions on construction and operation conditions; and restrictions related to cavity stability requirements such as mechanical strength.

The characteristics of high-level radioactive wastes are that they contain not only nuclides with a very high specific radioactivity and a relatively short half life, such as ^{90}Sr and ^{137}Cs, but also nuclides which have a relatively low specific radioactivity but have a long half life, such as ^{99}Tc and ^{237}Np. This means that because of these long-lived nuclides, the radioactive decay of vitrified wastes takes a considerably long period of time. As shown in Fig. 2.5, the time required for the radioactivity level of vitrified waste from 1 MTU of nuclear fuel to become the same as the radioactivity level of the uranium ore needed to manufacture fuel is of the order of tens of thousand years. When evaluating the safety functions of a geological disposal system, it is necessary to take into consideration many factors such as the long-term stability of the host rock. Furthermore, there is also a need to take uncertainty into consideration.

In Japan, research and development efforts have been continued for more than 30 years. In November, 1999, Japan Nuclear Cycle Development Institute (now JAEA) submitted to the Japan Atomic Energy Commission a technical report, “Technical Reliability of the Geological Disposal of High-level Radioactive Waste in Japan: Second Progress Report on Research and Development for Geological Disposal” [10] (“Second Progress Report”), integrating the R&D results accumulated by then. The report, which consists of the Project Overview Report, Supporting Report 1 (Geological Environment in Japan), Supporting Report 2 (Repository Engineering Technology), Supporting Report 3 (Safety Assessment of Geological Disposal Systems) and Supplementary Report (Background of Geological Disposal), indicated that geological disposal technology is reliable and is technically feasible in Japan. (These reports have been summarized into English version “H12: Project to Establish the Scientific and Technical Basis for HLW Disposal in Japan” (“H12 Report” in short).)

After submitting the Second Progress Report, Japan Nuclear Cycle Development Institute, as the second stage of the R&D process, set out to develop technologies for enhancing the reliability of geological disposal technology and refining safety assessment techniques by using the Mizunami Underground Research Laboratory (opened in April 2002) in Mizunami City, Gifu Prefecture, the Horonobe Underground Research Center (opened in April 2001) in Horonobe Town, Hokkaido, and the quantitative assessment radionuclide migration experimental facility at Tokai Village, Ibaraki Prefecture [23]. The Mizunami Underground Research Laboratory is engaged in R&D of technologies for safe and rational design, construction, maintenance and management of tunnels deep underground by conducting studies, for example, on the properties and distribution of faults and cracks in granite, the flow and chemical components of groundwater, and the strength of *in situ* rock. At Horonobe Underground Research Center, a large test boring base has been constructed in a sedimentary rock formation at a depth of about 250 m from the ground surface in order to further geoscientific research, enhance the reliability of disposal technologies including the verification of engineered barrier construction technology, and refine safety assessment methods. Similar research and development efforts have been continued in many other countries including the U.S., France, Sweden, Switzerland and Finland, and various exchanges are being made among these countries. In May 2001, Finland decided to construct a geological disposal facility at Olkiluoto, and in June 2009, Sweden decided to site a geological disposal facility at Östhammer.

In Japan, the siting process proceeds in three stages.

1. First stage—Selecting screening study areas: Literature and data research (literature research) is conducted, and the areas considered in the literature research are narrowed down to a smaller number of screening study areas.
2. Second stage—Selecting detailed study areas: Surface reconnaissance, boring, trench excavation and geophysical exploration are conducted, and detailed study areas are selected from the screening study areas.
3. Third stage—Selecting final disposal facility sites: Underground facilities are actually constructed, the physical and chemical properties of geological formations are investigated, and final disposal facility sites are selected from the detailed study areas.

As an institution charged with carrying out disposal, the Nuclear Waste Management Organization (NUMO) of Japan was established in October 2000, under the Specified Radioactive Waste Final Disposal Act. In December 2002, to select screening study areas, NUMO began to publicly seek feasibility study areas for high-level radioactive waste final disposal facility sites from among the municipalities in the country. NUMO has named this method an “open solicitation” approach. The method of publicly seeking candidate areas, which was then unprecedented in Japan for a large-scale project, was adopted on the basis of the idea that if a disposal project, which is highly public in nature and continues over a long period of time, is to be sited, it is necessary to respect the voluntariness of each municipality.

Besides, Finland was once thinking of transporting spent nuclear fuel to Russia. In 1994, however, Finland banned the transportation of spent fuel out of the country and abolished the condition that Olkiluoto (which was in those days thought to be a leading candidate site) be removed from the list of candidate sites (the situation was then called a “zero start”). This was when Finland started its siting process. Candidate sites were screened through *in situ* rock studies such as active fault studies conducted in many areas in the country, and safety assessment was conducted for the candidate sites thus selected. Olkiluoto, one of the candidate sites thus selected, was at first opposed to the proposed siting, but in January 2001, it decided to accept the project as a result of a vote within the municipality (20 votes in favor and 7 against) conducted separately from Parliamentary deliberations. Such a change in attitude resulted from not only the fact that the minimum requirement of ensuring safety was being met but also the fairness of the national government’s “zero start” policy. Another factor is that because nuclear power plants (Olkiluoto Unit 1 in 1978 and Unit 2 in 1982) and medium- and low-level radioactive waste disposal facilities (operated since 1992 by Teollisuuden Voima Oy, or TVO) had already been sited, the importance of the disposal project was understood, and there was a relationship of trust with the nuclear industry. Furthermore, the municipality had experience in developing a vision of the future of sited facilities under the initiative of citizens (supported by the national government and the implementing entity). Thus, besides the obvious requirement of ensuring safety, understanding and belief on the part of the citizens of the municipality concerned are needed because the project is a long-term one involving three generations.

In Japan, one of the greatest concerns related to geological disposal may be whether geological disposal is really possible in a country prone to earthquakes and other geological disturbances [5, 10, 23]. It is assumed in this chapter that formations including active faults are, as a general rule, avoided, and the absence of active faults in the space (about 2 km × 3 km) to be occupied by the proposed disposal facility is verified through geological surveys, geophysical exploration and excavation. It is nevertheless necessary, however, to assume the existence of active faults that cannot be detected through those surveys and investigations. JAEA’s approach is to assume a case where some of the engineered barriers protecting emplaced waste forms are crossed by a fault and to study possible behaviors of bentonite and overpacks due to fault activities. As a result of these studies, it has been confirmed both experimentally and analytically that although the bentonite layer is broken, the overpack is inclined but not broken. With respect to earthquake motion (seismic intensity), it is generally known that if a displacement of several centimeters occurs along a fault, aboveground facilities on the free surface of ground will be shaken intensely. This means that earthquake motion is smaller in the ground than at the ground surface. It is possible, however, that engineered barriers fail to function as expected if, for example, the state of contact between the bentonite and the overpack in an engineered barrier system changes temporarily. In the disposal system evaluation mentioned in the Second Progress Report, such irregular phenomena are taken into consideration, and it is verified that the radiation protection capability has a margin of safety even in the event of partial damage to the engineered barriers. In Japan also, it is possible to carry out disposal with a margin of safety by siting disposal facilities appropriately and using a combination of engineered and natural barriers in a mutually complementary manner.

This chapter has dealt with radioactive wastes from the viewpoint of radiation protection, but it is also important to protect the biosphere from the chemical toxicity of wastes. In Japan, JAEA conducted a study on toxic substances contained in TRU wastes [24]. In the study, a model for evaluating the migration of toxic substances contained in TRU wastes was developed for landfill disposal, concrete vault disposal and subsurface disposal, and limits for toxic substances at disposal facilities of each type were calculated. The evaluation method used is similar to the method used for the evaluation of the migration of radionuclides (see Chap. 7). When using this method, it is necessary to take chemical forms into consideration. Under the current legal system, radioactive materials to be regulated under the Act on the Regulation on Nuclear Source Material, Nuclear Fuel Material and Reactors are outside the scope of application of the environmental laws and regulations including the Environment Basic Act. After the management period of the disposal facilities ends and permission is granted for decommissioning, however, the environmental laws and regulations become applicable [24]. The report titled "Common Important Issues for the Safety Regulation of Radioactive Waste" [5] shows that the U.S. Environmental Protection Agency (EPA) has established concentration criteria, other than dose criteria, from the viewpoint of the regulation of groundwater contamination. In fact, the environmental protection criteria of the EPA have been applied to the Waste Isolation Pilot Plant (WIPP) in the U.S., which went into operation in 1993. The WIPP is a disposal facility for radioactive waste containing transuranium nuclides generated from military facilities by the reprocessing of spent fuel. In Japan, it is necessary to build disposal facilities designed by giving consideration to groundwater pollution after the end of the management period, when the waste disposed is released from regulation as radioactive wastes.

Exercises

1. Describe the role of natural barriers in geological disposal systems.
2. Verify that the volume of vitrified waste from the electricity consumed by one person in Japan during her or his lifetime (80 years) is about 0.1 liter if the ratio of nuclear power generation to total electricity generation is 1/3 and the burnup of nuclear fuel is 45,000 MWd/MTU. Assume here that the total annual power consumption (sales) is 919.5 TWh (2007), Japan's population is 128 million (2007), power generation efficiency is 33.4 %, the quantity of vitrified waste generated per ton of nuclear fuel is 1.25 forms/MTU, and the net volume of a vitrified waste form is 0.15 m^3.

References

1. US National Academy of Sciences/National Research Council, *The Disposal of Radioactive Wastes on Land* (US National Academy of Sciences/National Research Council, Washington, DC, 1957)
2. Nuclear Energy Agency of the Organisation for Economic Co-operation and Development, *Objectives, Concepts and Strategies for the Management of Radioactive Waste Arising from Nuclear Power Programmes* (Nuclear Energy Agency of the Organisation for Economic Co-operation and Development, Paris, 1977)
3. Svensk Kärnbränslehantering AB (Swedish Nuclear Fuel and Waste Management Co.). *Final Storage of Spent Nuclear Fuel: KBS-3*, SKBF/KBS Report KBS-3 (1983)
4. P. Witherspoon, G. Bodvarsson (eds.), *Geological Challenges in Radioactive Waste Isolation: Fourth Worldwide Review*, LBNL-59808 (Lawrence Berkeley National Laboratory, Berkeley, 2006).
5. Nuclear Safety Commission of Japan, *Common Important Issues for the Safety Regulation of Radioactive Waste*, June 10, 2004 (2004)
6. Nuclear Safety Commission of Japan, *Fundamental Principles on the Safety Review of Radioactive Waste Burial Facilities*, March 17, 1988 (Revised in part in March 29, 2001) (2001)
7. Japan Atomic Energy Commission Special Committee on Nuclear Power Back-end Measures, *Basic Principles on the Disposal of Low Level Radioactive Wastes that Exceed the Government Specified Limits* (Japan Atomic Energy Commission Special Committee on Nuclear Power Back-end Measures, Tokyo, 1998)
8. Atomic Energy Society of Japan, *AESJ Standard for Subsurface Disposal Safety Assessment Methods: 2008* (Atomic Energy Society of Japan, Tokyo, 2009)
9. K. Kato, T. Waki, N. Saito, F. Ono, T. Ohma, M. Ozaki, State of study for establishing the technical reliability of subsurface disposal. J. Nucl. Fuel Cycle Environ. **13**(1), 49–64 (2006)
10. Japan Nuclear Cycle Development Institute, *Technical Reliability of the Geological Disposal of High-Level Radioactive Waste in Japan – Second progress Report on Research and Development for Geological Disposal – Project Overview Report: Safety Assessment of Geological Disposal Systems*. JNC TN1400 99–20 (1999)
11. Nuclear Energy Agency of the Organisation for Economic Co-operation and Development, *Disposal of Radioactive Waste: An Overview of Principles Involved* (Nuclear Energy Agency of the Organization for Economic Cooperation and Development, Paris, 1982)
12. Japan Atomic Energy Commission, *Measures for Radioactive Waste* (Japan Atomic Energy Commission, Tokyo, 1976)
13. KASAM/SKN, *Ethical Aspects on Nuclear Waste: Some Salient Points Discussed at a Seminar on Ethical Action in the Face of Uncertainty in Stockholm*, Sweden, September 8–9, 1987, SKN Report 29 (1988)
14. S. Hirusawa, Geological disposal of high-level radioactive waste: a study on uncertainty and reliability. Kiho Enerugi Sogo Kogaku **27**(3), 101–116 (2004) (in Japanese)
15. US National Academy of Sciences/National Research Council, *Rethinking High-Level Radioactive Waste Disposal: A Position Statement of the Board on Radioactive Waste Management* (National Academy Press, Washington, DC, 1990)
16. Nuclear Energy Agency of the Organisation for Economic Co-operation and Development, *Post-Closure Safety Case for Geological Repositories: Nature and Purpose*, NEA No. 3679 (2004)
17. International Atomic Energy Agency, *Disposal of Radioactive Waste*, Specific Safety Requirements No. SSR-5 DS354 (2009)
18. I. Fujii, *Natural Reactors* (The University of Tokyo Press, Tokyo, 1985)
19. Radioactive Waste Management Funding and Research Center, *Reactors Created by Nature: From Birth to Disappearance and Clues to Successful Geological Management* (Radioactive Waste Management Funding and Research Center, Tokyo, 2005)

20. M. Kawanishi, Providing support to facilitate downstream projects: development of advanced technologies for transportation and storage of spent fuel and radioactive waste disposal. J. At. Energy Soc. Jpn. **52**(1), 21–27 (2010)
21. T. Kawata, What should fuel cycle experts and radioactive waste experts should do to ensure the sustainability of nuclear energy? 2. J. At. Energy Soc. Jpn. **52**(1), 28–32 (2010)
22. Federation of Electric Power Companies of Japan and Japan Nuclear Cycle Development Institute, *Second Progress Report for Research and Development for TRU Waste in Japan* (Federation of Electric Power Companies of Japan, Tokyo, 2005)
23. Japan Nuclear Cycle Development Institute, *Development of the Technical Knowledge Base for the Geological Disposal of HLW—2005 Report, Knowledge and Management of Geological Disposal Technology* (Japan Nuclear Cycle Development Institute, Tokai, 2005)
24. R. Kuroki, K. Takahashi, *A Study on Toxic Substances Contained in Radioactive Waste*, JAEA-Research 2007–082 (2008)

Chapter 7
Performance Assessment of the Radioactive Waste Disposal System

Joonhong Ahn and Shinichi Nakayama

7.1 Performance Assessment of the Disposal System

When geological disposal is planned, the first and the most fundamental question to be answered is: How safe is it? This comes not only from the regulatory authorities but also from the general public, waste generators and waste disposers. Performance assessment has been carried out over the years to answer this question.

The most common approach when considering safety is to separately consider safety during the operation period in which the repository receives wastes, and safety during the post-closure period. The former safety can be achieved and maintained in a similar manner as with other types of nuclear facilities, and there is accumulated experience in doing so. For safety of geological disposal, therefore, focus should be put mainly on the post-closure period. Waste to be disposed of deep underground includes long-lived radionuclides with a half life longer than 1 million years. The post-closure safety assessment period, therefore, is far longer than the history of human civilization.

For assessing long-term safety of geological disposal, the approach of analyzing system behavior by modeling it in accordance with scientific principles, expressing it mathematically and analyzing it by using a computer model is effective, just as is the case of dealing with many complex problems facing modern society (for example, global warming). As described in this chapter, performance assessment

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The nuclear regulatory system in Japan has been changed significantly after the Fukushima Daiichi Nuclear Power Station accident in March 2011. Descriptions in this chapter have been translated from the book originally published in Japanese before the accident, with minimal update where appropriate.

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tasks can be boiled down to this task. The basic structure for performance assessment had already been established by the 1980s in countries such as the U.S. and Sweden, and similar approaches have since been adopted in all countries planning geological disposal. In view of these circumstances, the International Atomic Energy Agency (IAEA) formulated an international framework jointly with the Nuclear Energy Agency of the Organisation for Economic Co-operation and Development (OECD/NEA) [1].

There are cases where besides "performance assessment," the term "safety assessment" is used. Performance assessment can be conducted for the entire geological disposal system or for a region or component of a system, for example, an engineered barrier. When an assessment is carried out to evaluate quantities specified in safety standards (for example, the maximum permissible exposure dose rate), performance assessment is often termed safety assessment because a safety judgment (regulatory) can be made according to the results of the performance assessment of the entire system. Performance indices other than safety indices, such as disposal cost, the area of the repository and proliferation resistance, can also be used for performance assessment. In this chapter, performance assessment and safety assessment may be considered synonymous because this chapter deals with performance assessment with respect to the safety of the entire system.

7.1.1 Purpose of Performance Assessment

Because performance assessment may be deemed synonymous with safety assessment as mentioned above, the purpose of performance assessment may be reworded as indicating assessment results related to long-term safety resulting from geological disposal. In performance assessment, it is necessary to determine indices and criteria in advance. For example, details such as who or what needs to be protected, how long they need to be protected and how accurate assessment results need to be are to be specified by indices, while the required level of safety is something to be dealt with by means of criteria because it is related to regulatory issues and public consensus building issues. The latter is discussed in Sect. 7.3.

As noted earlier, regulatory authorities set standards for the purpose of safety confirmation. Standards are determined according to social needs and by comparison with the safety and risks of other engineering systems in our society. Representative examples are cumulative releases of radioactivity from the repository in the Waste Isolation Pilot Plant (WIPP) in the U.S., exposure dose rates for the general public in the U.S. and other countries, and health hazard risks in Sweden. In the WIPP, the maximum permissible limit of cumulative releases of radioactivity in 10,000 years after closure is given for each nuclide as the ratio to the radioactivity inventory in the buried wastes. The method of specifying requirements in terms of the exposure dose rate has been adopted by many countries, and the limits thus specified range roughly from 0.1 to 1 mSv/y. IAEA recommends 0.3 mSv/y [1]. The limits at the Yucca Mountain repository in the U.S. are 0.15 mSv/y in the first 10,000 years after closure and 1 mSv/y in the term between 10,000 and 1 million years.

While the purpose of protecting future generations and the environment is common to all of the regulatory standards mentioned above, the regulatory system affects how repositories are to be installed. For example, if releases are regulated in terms of the cumulative release rate, the designer of a repository aims to take an approach of storing a large volume of waste in a large-scale repository if the isolation capability is high (groundwater-mediated migration is very slow). In fact, the WIPP is located in a salt formation, where the existence of groundwater can be ignored. If releases are regulated in terms of the maximum permissible exposure dose rate or risk, while isolation for a certain period of time is important, options such as locating a repository near a coastline can be considered because reduction in exposure dose rate due to slow releases from a repository or dispersive dilution in geological formations can be expected. Another option is to distribute wastes into a number of small repositories so that the quantity of wastes buried at each repository is small.

Performance assessment is expected to play a number of different roles when a repository goes into operation and at different stages until closure. At stages in which disposal sites are yet to be determined, performance assessment is used as a feasibility evaluation method for determining, for example, whether geological disposal is feasible under the conditions in a particular country and, if feasible, what kind of specifications should be used. In such cases, since geological conditions are yet to be identified, performance assessment is made by categorizing geological conditions in the country and designing engineered barriers that are thought to be suitable for those conditions. The so-called “H3 Report” (1991 Report) [2] and “H12 Report” (2000 Report) [3] in Japan are considered to be the results of such R&D efforts. As can be imagined easily, there are many unknown and undetermined things at this stage, and the results obtained contain significant uncertainties. The obtained results, therefore, are carefully examined, and evaluation is repeated by improving the data and models used. Non-site-specific performance assessment like this is called generic assessment.

If it has been concluded as a result of a feasibility study that geological disposal is possible and if a policy decision has been made to go with geological disposal, the next step is site selection. In site selection, a common approach is to narrow down a list of candidate sites. In this case, too, performance assessment provides most of the knowledge needed. At this stage of performance assessment, a greater amount of knowledge about actual geological formations should be available than at the stage of the feasibility study. Performance assessment, therefore, can be made on the basis of more concrete and specific knowledge.

When a site has been selected as a result of the site selection process, the waste disposer must show to the regulatory authority that the site is safe and obtain a permit and/or license for the disposal project. Performance assessment at this stage is most detailed.

License application is to be made by stages. The first to be obtained is one for the construction of an underground disposal facility. Subsequently, a permit needs to be obtained for any change. Very likely the highest hurdle to be cleared is for the closure of the repository after completion of waste emplacement. The regulatory authority makes pre-closure performance checks by conducting such as monitoring to see if, for example, repository performance has been achieved as expected. License based on detailed performance confirmation needs to be issued to close the repository.

Thus, performance assessment functions as a playmaker and a compass at each stage of geological disposal.

7.1.2 Content and Method of Performance Assessment

As noted above, the purpose and use of performance assessment differs from stage to stage. Then, specific steps in performance assessment can be summarized into the four stages shown below. These stages may proceed sequentially, but in reality they often proceed concurrently and iteratively.

1. Definition of disposal system details
2. Identification of events that are likely to occur after closure and affect the safety of the repository and scenario development
3. Development of models (system level and event/barrier level) and computer codes (including modeling of uncertainty)
4. Calculation of performance indices and comparison with safety regulations

These are explained next.

7.1.2.1 Definition of System Details

As mentioned in Chap. 6, the disposal system can be divided into three major components: waste forms; repository structures functioning as engineered barriers; and the geological environment functioning as a natural barrier. R&D often takes place in these three areas separately. When conducting a performance assessment, however, it is necessary to integrate information from all of these areas.

Some of the most important information concerning waste forms deals with the content, or inventory, of radionuclides. It is also necessary to know the physical and chemical characteristics of the substances (matrix) that carry the radionuclides. For example, heat conductivity, density, matrix structure (e.g., crystalline or amorphous) and the distribution of nuclides in the matrix are essential to characterize the waste form. Also chemical reactions of the waste form with groundwater and with corroded and intact metal container, which determine the long-term evolution of materials in this region, must be understood.

Engineered barriers include backfill materials and buffer materials as principal components. As mentioned in Chap. 6, buffers are added to keep emplaced waste forms in a stable environment over a long period of time. The composition of the groundwater that has passed through the buffer, corroded the metal container and reached the waste form is likely to differ considerably from those of the groundwater that exists in the original underground environment. The groundwater compositions are also affected by dissolved waste matrix and radionuclides. Radiation can greatly change environmental conditions through, for example, the radiolysis of water. Nuclides released from waste forms migrate in the engineered barriers. The reaction of these nuclides with the buffers and groundwater is also important.

The geological environment is an important region affecting the condition of engineered barriers. The geological environment can also be relied on as the last barrier to isolate radionuclides from the biosphere. It is necessary to have information concerning groundwater hydrology, chemical species dissolved in groundwater, and the reaction between groundwater and rock when nuclides migrate in the geological environment.

If the exposure dose rate is specified as a performance index, information on radionuclide migration in the biosphere is needed. It is necessary to define in advance critical groups who can be exposed through intake, define pathways leading to such exposure, and identify migration mechanisms in those pathways and various parameters.

7.1.2.2 Identification of Events Likely to Affect Safety and Scenario Development

It is not possible to analyze all future events. Events considered in performance assessment are only a small subset of all possible events. Such a subset of events must be selected carefully so that they represent future states of the repository under consideration. These are called scenarios. For the purpose of performance assessment, repository behavior is often described by using a number of representative scenarios.

It has become a common understanding that systematically developing an optimum set of scenarios that exhaustively cover all of numerous possible events is important for highly reliable performance assessment. There are a number of methods [4] to do that for this purpose. First of all, scenarios must be set so that compliance to regulatory requirements can be demonstrated. For example, if the exposure dose rate is required to be assessed by the regulation, it is necessary to develop scenarios by focusing on events that can affect future exposure of the general public.

Although it is not possible to make a list of all possible events in scenario development, it is essential, as the next best thing that can be achieved, to keep a record of the process of scenario development, in which events have been systematically and exhaustively investigated and listed. Such a record becomes not only an important document for license reviews but also basic information needed when unlisted events are newly found.

Usually, the first step in scenario development is to list conceivable events. These events are classified into Features, Events and Processes (FEPs), and the list thus made is called a FEP list. The differences among the three categories are as follows. For example, according to the U.S. Nuclear Regulatory Commission (NRC), Feature is an object, structure or condition that has a potential to affect disposal system performance; Event is a natural or anthropogenic phenomenon that has a potential to affect disposal system performance and that occurs during an interval that is short compared to the period of performance; and Process is a natural or anthropogenic phenomenon that has a potential to affect disposal system performance and that operates during all or a significant part of the period of performance. In reality, however, it is rarely necessary to strictly distinguish between these. In fact, usually, these are collectively called FEPs.

Scenario development has continued for more than 30 years in a number of countries, and OECD/NEA put together and published the scenarios thus developed [5]. Thus, international comparison of scenarios is useful in developing a closer-to-perfect list of FEPs.

When a provisional list of FEPs has been completed, the next step is to screen FEPs on the basis of that list according to such factors as the probability of occurrence of each FEP and the degree of influence on performance. FEPs thus selected are used to develop scenarios. At this stage, it is important to keep a record of the screening conditions and the FEPs screened out. For example, according to the Standard 40 CFR 197 set by the US Environmental Protection Agency, the exclusion criterion level is 10^{-8} per year or lower in occurrence probability or the level at which the results of performance assessment would not change significantly by the omission of the FEP under consideration.

A method for enhancing the reliability of the FEP list is the preparation of an influence diagram or an interaction matrix (see for example, Reference [6]). Another useful approach is to classify the listed FEPs into a number of groups and examine their relationship.

7.1.2.3 Development of Models and Computer Codes

When scenarios are ready, models are constructed on the basis of the scenarios, and computer codes are created. The basic storylines are engineered barrier deterioration, nuclide release from waste forms, nuclide migration in engineered and natural barriers, nuclide migration to the biosphere, and human intake. FEPs, particularly in an engineered barrier, are in a coupled state, where FEPs affect one another. For example, bentonite, which is used as a buffer material, swells from groundwater infiltration. The degree of swelling is affected by not only the type of bentonite but also the dissolved components of groundwater. Swelling affects groundwater flow in the engineered barrier and the excavation-damaged zone surrounding them.

Metallic container corrosion affects the internal environment of the engineered barrier and the swelling of bentonite by causing the concentration of oxygen in groundwater to decrease and causing hydrogen to be generated. Corrosion products expand so as to mechanically apply pressure to bentonite because the density of corrosion products is lower than that of the original metal. The radionuclides, particularly alpha-emitting radionuclides, released as a result of the dissolution of the matrix in the waste forms cause the radiolysis of water so as to affect the environment. Thus, the inner part of the engineered barrier is in a coupled state, where hydrological, chemical, mechanical and radiochemical processes affect one another. It is also necessary to take the effect of temperature into consideration because considerable heat generation from waste forms continues over a period of more than 1,000 years after the start of burial of waste forms.

Recent years has seen remarkable progress in the method of solving simultaneous governing equations. In this commonly used numerical approach, various coupled processes are solved separately and then connected again after they are solved. For example, by assuming that a chemical reaction proceeds with sufficient speed at each time step, the effects of chemical processes in the swelling of bentonite are modeled separately.

Another type of decoupling is made spatially. Since the conditions in an engineered barrier region, a natural barrier region and the biosphere differ considerably, radionuclide migration analysis is conducted separately in each of these regions, and the results are connected subsequently to evaluate the entire system. Although no problem arises as long as the continuity conditions are satisfied at each of these interfaces, there are cases where the continuity of radionuclide concentration in the water phase is lost. Care needs to be taken, therefore, depending on the performance indices used [7].

7.1.2.4 Calculation of Performance Indices and Comparison with Safety Regulations

Experimental knowledge is essential for scenario development and model development for the geological disposal system. For the purpose of acquiring data and making *in situ* observations it is important to provide underground research facilities prior to or concurrent with repository construction.

The maximum time scale of knowledge that can be gained through experiments or *in situ* observation is at most about 10 years, which is much shorter than the length of time considered in connection with disposal safety evaluation. It is possible, however, to model long-term processes by deepening fundamental mechanistic understanding. Models used to deepen theoretical understanding in such cases (called process models or detailed models) need to be distinguished from models used to calculate performance indices. Model development for performance assessment can be understood by considering a hierarchical structure as shown in Fig. 7.1 [8].

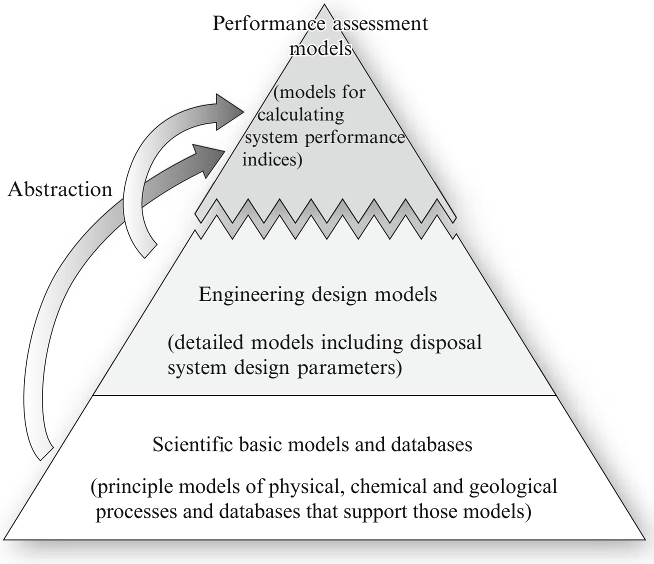


Fig. 7.1 Hierarchical structure illustrating performance assessment model development

The ultimate goal of performance assessment is to calculate performance indices specified in regulations. Because of the huge computing load, it is practically impossible to achieve that goal by simply joining together detailed models. Even if an enhancement in computing power has made it possible to do that, it is still difficult to answer questions regarding overall system behavior on a case-by-case basis (e.g., “What is the principal factor affecting system performance?”). If it takes several hours to several days to obtain a solution for a single case, it is not possible to examine many cases exhaustively. The probabilistic uncertainty analysis described in the next section poses similar difficulties. The approach to be taken, therefore, is to identify and simplify mechanisms and parameters that significantly affect performance indices on the basis of the knowledge gained from detailed models and develop a performance assessment model integrating those mechanisms and parameters. Conservative assumption is a widely used approach employed to convert detailed models into a performance assessment model. Conservative assumption is the assumption made so that the safety of a repository is underestimated. The idea is that if it can be shown that regulatory requirements can be met even if conservative assumptions are made with respect to safety, then it can be shown that the disposal system under consideration meets safety standards with a sufficient margin of safety. In some cases, rough sensitivity analyses are carried out by using detailed models, and detailed mechanisms that will not significantly affect performance are incorporated into the performance assessment model with simplified empirical approximations or even ignored. By using the performance assessment models and codes thus obtained, performance indices are evaluated, and, finally through comparison with the safety standards, the safety of the disposal system is evaluated. In spite of conservative assumptions, the results of performance assessments of various disposal facility concepts that have been shown in each country indicate that geological disposal can achieve a level of safety higher by several orders of magnitude than the level required by the safety standards. This is the technical basis for the present international consensus that geological disposal is safe.

Performance assessment is carried out within the framework of the scientific and technological knowledge of the time. Consequently, unknown areas at that stage (for example, the geological environment cannot be identified through generic assessment) are left unsolved by, for example, introducing conservative assumptions. There is still concern, therefore, with respect to some unanswered questions about, for example, whether conservative assumptions are really conservative and whether system behavior can be accurately reproduced when coupled conditions are separated and joined together again. It is a common understanding, therefore, that performance assessment should be carried out repeatedly at different stages of geological disposal development. It is important to study in detail and document how performance assessment results have changed through repetitive performance assessments and which part of the performance assessment model has caused such changes. This not only helps in understanding system behavior but also facilitates license reviews conducted by regulatory authorities in the future.

7.1.3 Treatment of Uncertainty

Performance assessment is accompanied by uncertainty due to various causes. Such uncertainty cannot be eliminated no matter how many advancements are made in science and technology and how many times performance assessments are repeated. Chapman and McCombie [9] investigated causes of uncertainty and classified uncertainty into four types: (1) system uncertainty due to incomplete understanding of the disposal system; (2) scenario uncertainty due to incompleteness of the scenarios incorporated into performance assessment; (3) model uncertainty due to the choice of models incorporated into performance assessment; and (4) parameter uncertainty associated with each parameter. Helton and Marietta [10] classified uncertainty into aleatory uncertainty and epistemic uncertainty in their study on uncertainty from a structural viewpoint. Aleatory uncertainty is the kind of uncertainty that cannot be eliminated no matter how many R&D results are accumulated. An example is uncertainty due to unpredictability of when and where an earthquake will occur and how strong a future earthquake will be. Epistemic uncertainty, which arises from a lack of knowledge, is the kind of uncertainty that can be reduced by increasing the amount of knowledge and information available. For example, by increasing the number of observation boreholes at and around the disposal site, the amount of information available on the host rock increases, and uncertainty decreases in inverse proportion to the amount of such information. In reality, however, even in such cases, usually a considerable degree of uncertainty remains.

Methods of handling uncertainty in performance assessment can be broadly classified into two approaches. One of them is the probabilistic approach, in which the uncertainty about parameters in a performance assessment model is expressed with a probability density function, and the uncertainty about the performance indices arising from that is also expressed with a probability density function. The other is the conservative approach, in which model and parameter values are selected so that assessment values given by performance indices are pessimistic.

In the former approach, the structure of uncertainty can be expressed quantitatively. When two or more sites or design options are compared, quantitative information is given as to what degree the sites or options differ significantly. The amount of information, therefore, available for public decision making is large. In the case of parameter uncertainty, parameter values are sampled on the basis of the probability density function defined for each parameter, often by using the Monte Carlo method, and performance assessment indices are calculated by using the parameter values thus obtained. By repeating this procedure, statistical distributions of the performance assessment indices are obtained. The latter approach (conservative approach) excels in simplicity of logic when explaining results. Because safety standards are often indicated in the form of limit values such as upper limit values, a properly constructed conservative model provides assessment results with a solid logical foundation. Examination of examples of performance assessments carried out in different countries reveals that these two approaches have been used in all performance assessments. Since various conservative assumptions are incorporated into performance assessment models at their development stage, the results obtained need to be interpreted carefully even if they have been obtained by the probabilistic approach.

For system uncertainty, scenario uncertainty and model uncertainty, different deterministic model systems are constructed and compared. For each model system, parameter uncertainty is estimated by a method similar to the method described above. As a result, assessment results reflecting parameter uncertainty are obtained for multiple scenarios and models. Assessment results may be weighted by defining the probability of occurrence of scenarios and models so as to obtain final assessment results; or the results from multiple scenarios and models may be indicated separately as final results of assessment.

7.2 Performance Assessment Models

7.2.1 Groundwater Hydrology and Geochemical Environments

7.2.1.1 Groundwater Movement

The most important scenario in the performance assessment of the radioactive waste disposal system is the groundwater migration scenario. It is generally thought that groundwater migration is sure to occur. The probability of occurrence, therefore, is 1. Future human exposure dose rates are estimated by mathematically expressing the radionuclide transport from the dissolution of radionuclides from wastes buried at the underground repository through their movement along groundwater pathways leading to the biosphere. As the first step, therefore, it is necessary to obtain information on groundwater hydrology at and near the repository, particularly groundwater pathways and groundwater flow velocity.

Groundwater flows from higher potential regions to lower potential regions, that is, from ridges to valleys, from inland regions to coastal regions, passing through voids and cracks in rocks. This movement is called gravity-driven flow. The driving force for groundwater flow is this topographic gradient. A formula expressing water flow driven by the topographic gradient is Darcy's law.

$$Q = AKi \tag{7.1}$$

This relation was originally derived experimentally for sand formations, a typical porous medium. The discharge rate Q [m^3/s] through a sand layer is proportional to the cross-sectional area of flow A [m^2] and the hydraulic head Δh [m] of the fluid between the point of inflow and the point of outflow in the sand layer and is inversely proportional to the thickness s [m] of the sand layer. In Eq. (7.1), the hydraulic head per unit distance, $\Delta h/s$, is expressed with the hydraulic gradient i. The proportionality constant K [m/s] in this case is the hydraulic conductivity. Although K has the unit "m/s," it is not a value expressing velocity. The discharge rate per unit cross-sectional area, Q/A, is denoted as q. This q [m/s] is called the Darcy velocity. It is often called flow velocity because it has the unit "m/s." Strictly speaking, however, it is flux instead of flow velocity. The actual flow velocity, or the pore velocity of groundwater, is a value obtained by dividing q [m/s] by porosity ε.

Porous media such as sedimentary rock have pores between particles, and groundwater moves through these pores. These pores are called matrix pores. The hydraulic conductivity K for a porous medium can be expressed with the density ρ [kg/m^3] of water, the kinematic viscosity μ [kg/m s], the intrinsic permeability k [m^2] and gravitational acceleration g [m/s^2], as

$$K = \rho g k/\mu \tag{7.2}$$

Since the permeability of porous rock is proportional to the square of the mean particle diameter d [m], the intrinsic permeability k can be calculated by using the Kozeny–Carmen equation;

$$k = \frac{\varepsilon^3}{(1-\varepsilon)^2} \cdot \frac{d^2}{180} \tag{7.3}$$

In hard rock such as granite, groundwater moves through fractures because the matrix is highly compacted. For such fractures, aquifers and storage layers, the transmissibility T [m^2/s], instead of the hydraulic conductivity, is used. A fracture is modeled as two parallel plates placed apart at a fixed aperture (parallel plate model). Letting b [m] represent the aperture, then, T is the product of b and the hydraulic conductivity K as in Eq. (7.4).

$$T = Kb \tag{7.4}$$

With the extension w [m] of the fracture, the volumetric water flow rate Q can be written by Eq. (7.1) as $Q = Kbwi = Twi$. If the fracture is assumed to be two parallel plates, the hydraulic conductivity K in the fracture can be expressed as $K = \rho g b^2/12\mu$. Hence, the volumetric water flow rate Q in the fracture is proportional to the cube of the aperture b. This is called the cubic law.

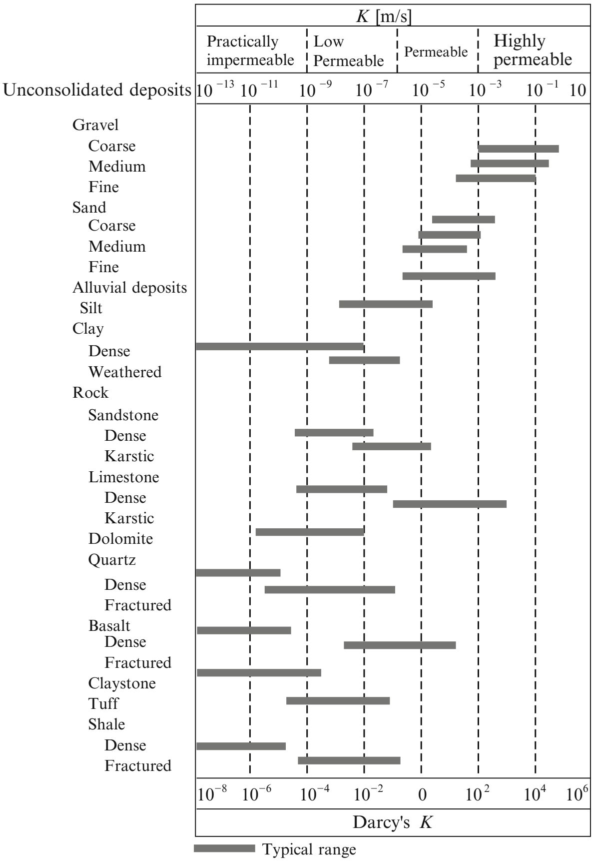
In limestone, karstic fractures formed by the dissolution of lime over a long period of time can be seen, and limestone having such fractures may also be thought of as fractured rock. In general, however, fractures in a rock mass cannot necessarily be clearly classified either as matrix pores or fractures. In sedimentary rock, there are also fractures other than pores between particles.

Possible driving forces for groundwater movement other than the hydraulic gradient due to topography include heat convection and density-driven flow. Heat convection is a flow that occurs when there is an underground heat source. A known example is deep fluid under the influence of the heat of magma. At the geological disposal facility, radionuclides decay in the vitrified waste forms before they are released into the surrounding rock so that heat generation becomes insignificant. There is no need, therefore, to take into consideration the occurrence of heat convection. Heat generation in the initial stage after vitrified waste forms are created is due to the decay heat of radionuclides with a relatively short half life, such as ^{90}Sr (29.1 y) and ^{137}Cs (30.2 y). Engineered barriers used in geological disposal are capable of containing those nuclides for several hundred years until those nuclides decay sufficiently.

Density-driven flow is a flow that occurs when two fluids of different densities come into contact, driven by the density difference between the two fluids. In a coastal area, density-driven flow occurs because of the density difference between inland water (freshwater) and seawater (saline water). On the inland water side of the fresh-saline water boundary, upflow occurs toward the coastal zone, and on the seawater side, downflow occurs toward the deeper side of the ground. When constructing the disposal facility in a coastal area, it is necessary to take this into consideration.

The hydraulic conductivity K is highly dependent on the physical properties of rock. Typical values for different media are shown in Fig. 7.2. The values range from about 10^{-3} to 10^{-1} m/s for gravel and from about 10^{-5} to 10^{-3} m/s for sand. The values for shale and basalt, both of which are densely consolidated, are as small as about 10^{-8} m/s or less.

Fig. 7.2 Ranges of hydraulic conductivity for major geological media [11]



In the *in situ* permeability tests conducted by Japan Nuclear Fuel Ltd. (JNFL) at boreholes drilled in the southern part of Shimokita Peninsula for the purpose of surveys for subsurface disposal in Japan, a hydraulic conductivity of 9.9×10^{-8} m/s was obtained from a sand formation interspersed with pumice at a depth of 80 m, and an average hydraulic conductivity of 1.9×10^{-8} m/s was obtained from a pumice tuff formation [12].

In deeper parts of the ground, the influence of surface landforms is relatively small, and the hydraulic gradient, which is the driving force for groundwater flow, tends to be small at greater depths. It is generally believed that at depths of several hundred meters from the ground surface, where geological disposal facilities are to be located, groundwater flow velocity is about several tens of centimeters to several meters per year. These very low flow velocities are by no means in the range that can be observed in the experiment conducted on sand formations by Darcy. Laboratory measurement methods for hydraulic conductivity measurement include the constant head method, falling head method, transient pulse method, and the oscillation method. Field measurement methods include the pumping method and the injection method. The hydraulic conductivity measurement method to be used is selected according to the magnitude of the hydraulic conductivity to be measured. In general, unignorable errors arise in the measurement of small hydraulic conductivity values. A study has been conducted on measurement errors accompanying laboratory measurement methods [13].

Basically, groundwater movement is viewed as a spatially continuous cycle consisting of a recharge zone where rainwater and snow melt seeps into the ground from the ground surface, a transmission zone where groundwater moves, and a discharge zone where groundwater flows out at the ground surface into rivers and lakes. In groundwater hydrology, the groundwater table plays an important role. The groundwater table is the plane in which both groundwater and air exist. The layers overlying the groundwater table are unsaturated layers, and the layers underlying it are saturated layers. The depth of the groundwater table is roughly determined by the relationship between precipitation and permeability. In rainy regions like Japan, the groundwater table is formed at relatively shallow depths of several meters to more than 10 m below ground surface. Among the radioactive waste disposal concepts conceived for use in Japan, landfill disposal and concrete vault disposal facilities are to be located in unsaturated formations. Subsurface disposal facilities, which are said to be constructed at depths of 50–100 m, and geological disposal facilities, which are to be constructed at depths greater than 300 m, are to be located in saturated formations. The Yucca Mountain repository was planned to be constructed in a desert area in Nevada is located at a depth of 300 m, and the groundwater table is located below that level. The radionuclide migration medium to be considered in performance assessment, therefore, is unsaturated formations as in the case of landfill disposal and concrete vault disposal in Japan. In the performance assessment of the Yucca Mountain geological repository, a radionuclide migration analysis was conducted by assuming gas–liquid two-phase flows.

In order to obtain information on groundwater hydrology at great depths as in the case of geological disposal in Japan, it is necessary to analyze the groundwater flow system extending over a large area such as an area including the disposal facility between faults or an area surrounded by ridges, instead of, a repository-scale (e.g., 2 km × 3 km) area. A regional groundwater flow analysis deals with a very long time scale. In the analysis, therefore, it is necessary to predict long-term groundwater movement by incorporating the prediction of topographic changes and sea level changes. It is also necessary to set boundary conditions for hydrological analyses for a smaller, repository-scale area. Developing a method for such analysis is by no means easy. The reason is that available data on geological and hydrogeological structure are limited. Boreholes reaching depths of several hundred meters below ground surface, which would provide high-quality data, are rare, and verifying computer codes to be used for analysis is very difficult to achieve. Research on large-scale groundwater flow is a theme in the field of earth science, and advances in the coming years are hoped for.

7.2.1.2 Geochemistry of Groundwater

The chemical properties of elements are primarily determined by valence, which is governed mainly by redox potential E_h. The chemical species of elements are determined by hydrogen ion concentration (usually expressed as $\mathrm{pH} = -\log [\mathrm{H}^+]$) and chemical reactions with various coexisting ions. When selecting data to be used for modeling of phenomena dealt with in the performance assessment of the disposal system such as the dissolution of elements, reactions with minerals and rocks, diffusion and corrosion of metals, it is necessary to have knowledge of geochemical characteristics of groundwater such as pH, redox potential, and composition.

The geochemical characteristics of groundwater are the results of the reactions of rainwater or seawater with soil or rock occurring over a very long period of time. As depth increases, groundwater of precipitation origin tends to change from the Na^+–$Ca^{2+}(Mg^{2+})$–HCO_3^- type to the Na^+–HCO_3^- type because $Ca^{2+}(Mg^{2+})$ decreases, and pH tends to change from neutral in the surface layer to mildly alkaline. Groundwater of seawater origin is rich in Na^+ and Cl^-, both of which are principal components of seawater.

The composition of groundwater of precipitation origin is formed as follows. Precipitation contains very small amounts of dissolved chemical species of aerosol origin such as Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, SO_4^{2-} and NO_3^-. Groundwater of precipitation origin also contains atmospheric components such as N_2, O_2, Ar and CO_2. In the surface layer, with the progression of organic matter decomposition by microorganisms in soil and decomposition by sedimentary bacteria, dissolved oxygen decreases, and concurrently CO_2 is produced so that it is dissolved into the soil and carbonate ion concentration increases. The partial pressure of CO_2 produced by organic matter decomposition is 10–100 times higher than that of the atmosphere. Surface layer water thus produced reacts with silicate minerals or carbonate minerals, and Na^+ and Ca^{2+} concentrations increase. As a result, Na^+–$Ca^{2+}(Mg^{2+})$–HCO_3^- type initial shallow groundwater is formed.

At depths greater than several hundred meters below ground surface, dissolved oxygen in water is consumed, and bacteria carry out the reduction of NO_3^- to N_2, the reduction of SO_4^{2-} to H_2S, and the fermentation of sedimentary organic matter. As a result of these reactions, HCO_3^- and Cl^- concentrations in groundwater increase relatively. Cations react with clay minerals, and Na^+ and Ca^{2+} exchange ions so that Na^+ concentration in the groundwater gradually increases and groundwater of Na^+-HCO_3^- type is produced.

Unlike water quality data, reliable data on redox potential at great depths are scarce. In the subsurface disposal survey introduced earlier, the pH and redox potential of sandstone formations interspersed with pumice at a depth of about 100 m were 8.9 to 9.5 and -224 to -24 mV, respectively, and those of pumice tuff formations at the same depth were 8.3 to 8.7 and -106 to 79 mV [12]. In the sedimentary rock formations at Horonobe in Hokkaido and Tono in Gifu Prefecture and in the granite formations at Tono, a reducing condition was observed at depths around 500 m [14]. Data on the oxidation and reduction of groundwater are scarce partly because there had been no need for such research until the concept of geological disposal was proposed and partly because groundwater sampling with minimal chemical disturbance (for example, improved boring designed to prevent the intrusion of water containing air or oxygen) was technologically difficult to achieve. The development of the technology to measure pH and redox potential without causing contamination or degassing of dissolved gas during boring is nearing completion [15, 16].

It is generally believed that underground oxidation and reduction conditions are created by the dissolution and sedimentation of iron-bearing minerals such as biotite and sulfide minerals such as pyrite. In the granite formations at Tono, redox potential is determined by Fe^{2+} and Fe^{3+} of biotite origin at depths of 180–330 m and chemical species containing sulfur such as $SO_4{}^{2-}$ and HS^- at depths of 500–1,000 m [15]. In the underground environment, about 10^4–10^6 cells of microorganisms also exist in each cubic milliliter of groundwater, and it has been found that these microorganisms contribute to the creation of the oxidation–reduction environment by causing the decomposition of organic matter in the underground environment and reactions such as nitrite reduction, iron reduction and sulfate reduction. It used to be believed that the reducing condition at great depths was the result of the reaction of minerals (i.e., inorganic matter). Research on microorganisms at great depths in recent years is revealing that microorganisms are playing a significant role in creating the environment of the earth's interior [17].

Thus, deep underground constitutes a low-oxygen reducing environment where the corrosion and dissolution of metals are slow. These geochemical characteristics of deep groundwater provide a technical basis for believing that geological disposal is valid, together with very slow movement of groundwater which mediates underground mass transport. It is therefore important to estimate and confirm that the underground environment of the planned repository site is reducing and that the reducing environment will be restored after the site is backfilled even if the site is exposed to the air during repository construction. It is thought likely that because of huge quantities of iron ions, sulfur compounds, and microorganisms, the oxygen that has entered the repository during construction will be gradually consumed so that the repository environment will eventually become reducing again. Assessment to determine how long that process will take, including a method of such assessment, is one of the areas for further study.

7.2.2 Source Term Model

Radioactive wastes buried at the repository are the source of radionuclides that migrate into groundwater. A source term is a mathematical model used in the performance assessment of a disposal system to express the starting point for the migration of radionuclides from the repository (engineered barrier) to rock or soil (natural barrier).

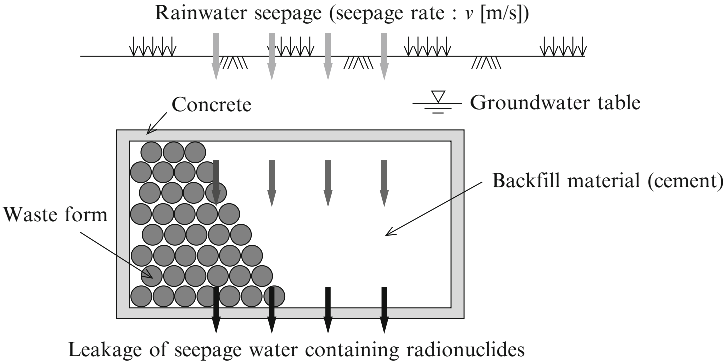


Fig. 7.3 Source term model of concrete vault disposal

Every disposal facility at which waste forms are buried consists of multiple barriers in order to prevent the intrusion of groundwater and the movement of radionuclides. Usually, wastes are packed in containers or solidified. For example, liquid waste and spent resin are mixed together and solidified in metal drums or, as is the case with the vitrified waste forms of high-level radioactive wastes, contained in canisters and enclosed in an overpack. In a source term model, it is not possible, however, to completely model various phenomena that occur during the process in which groundwater passes through multiple barriers, enters the repository, comes into contact with waste forms, dissolves radionuclides, and moves out of the repository (see Sect. 7.1.3). Modeling is carried out, therefore, by combining the main phenomena thought to be occurring in the repository structure.

Figure 7.3 illustrates a concrete vault repository, a kind of “near-surface” disposal facility for low-level radioactive wastes. A typical concrete vault measures about 40 m (length) × 40 m (width) × 5 m (depth). A number of assumptions are made for the purpose of modeling. As shown in the illustration, waste forms are stacked in the vault, and the space between them is filled with backfill material (cement in the example shown). In the model described below, however, the vault as a whole is regarded as a rectangular mass made of a homogeneous mixture of radioactive waste and backfill material. In other words, the waste form drums, the cement used to backfill the void space in the drums, and the concrete used as the exterior walls of the vault are not modeled individually. This is an extremely simplified and conservative assumption in a sense that it completely ignores the barrier capability of these materials. A state like this is thought to correspond to a complete loss of the barrier function that can occur in the far distant future when the constituent materials have deteriorated. In safety assessment, a condition like this is assumed immediately after the closure of the repository for conservativeness. This system is the same as that assumed when calculating clearance levels in Chap. 4 (see also Chap. 8).

It is assumed that rainwater that has fallen directly above the vault seeps into the vault at a velocity v [m/s]. This seepage water leaches out radionuclides in the vault and migrates from the bottom of the vault toward the natural barrier. Changes in the homogenized radionuclide concentration in the vault can be expressed as

$$\frac{dC_s}{dt} = -\left(\lambda + \frac{v}{L\theta_s R_s}\right)C_s \tag{7.5}$$

where

C_s: radionuclide concentration in the water phase in the vault [Bq/m^3],
λ: radioactive decay constant [1/s],
L: vault height (thickness) [m], and
θ_s: porosity of the mixture of backfill material and waste in the vault (ratio of pores in unit volume).

The subscript s denotes a source term. The decay chain is not taken into consideration.

In the equation, R_s $(= 1 + \rho_{b,s}K_{d,s}/\theta_s)$ is called the retardation factor, which includes the sorption distribution coefficient $K_{d,s}$ [m^3/kg]. The coefficient $K_{d,s}$ expresses the strength of radionuclide sorption to the solid phase in the vault. $\rho_{b,s}$ [kg/m^3] is the bulk density of the mixture (unit weight). The velocity of the radionuclide transport in the medium (i.e., in the vault, in this case) is reduced by $1/R_s$ compared to the seepage water velocity. In the equation, t [s] is the elapsed time counted since the time point when the vault is closed after waste form emplacement and backfilling are completed. Equation (7.5) governs the change in the radionuclide concentration in the water phase in the vault. In this model, the vault concrete wall is homogenized with other materials in the vault. Therefore, the radionuclide concentration in the water flowing out from the bottom of the vault to the natural barrier is given as C_s [Bq/m^3]. This is the source term.

As shown in Fig. 7.3, the concrete vault is located below the groundwater table. Since the pores below the groundwater table are saturated with water, it is assumed for Eq. (7.5) that the pores in the vault are saturated with seepage water. Pores in the ground above the groundwater table are not necessarily filled, or saturated, with groundwater. In this case, the degree of saturation is incorporated into the equation as a parameter.

The sorption distribution coefficient K_d is a very important parameter in performance assessment. If solid and water phases coexist in the system, part of the solute dissolved in the water is adsorbed onto the solid surfaces or absorbed by the solids. Adsorption and absorption are collectively termed sorption. The sorption distribution coefficient K_d is defined as the ratio at an equilibrium state between the quantity adsorbed onto the solids of unit weight [mol/kg solid] and the quantity left in the water phase of unit volume, or the water phase concentration [mol/m^3 water]. The sorption distribution coefficient K_d is 0 if the solid phase does not sorb the radionuclide of interest in the water phase. In this case, the retardation factor R_s becomes unity, meaning that the radionuclide is transported at the same speed as the

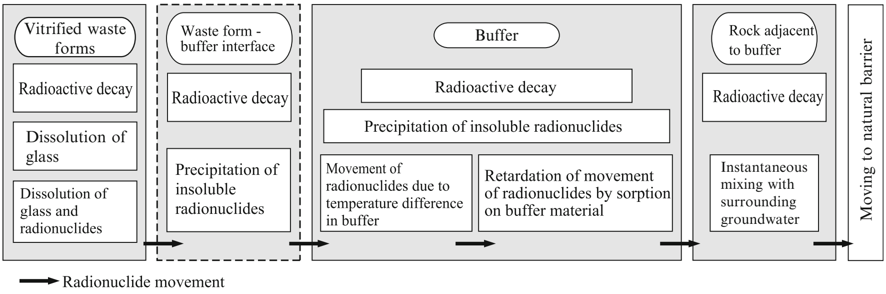


Fig. 7.4 Concept of radionuclide transport in engineered barrier at the geological repository [18]

groundwater flow. If the water flow is sufficiently slow compared with rates of chemical reactions with solutes and minerals, chemical equilibrium is considered to have reached at each local point. If this is the case, detailed reaction kinetics models are not necessary. The sorption distribution coefficient K_d is a phenomenological parameter, and can be experimentally measured for a given combination of the solids (rock, mineral, soil) and chemical species. A theoretical study has been conducted concerning adsorption (Sect. 10.3).

The source term in the performance assessment of geological disposal includes the migration of radionuclides through the buffer to the surrounding host rock. The geological disposal facility (repository) consists of the vitrified waste forms, overpack and bentonite buffer, each of which is expected to function as a barrier by confining radionuclides, delaying water infiltration, and retarding radionuclide transport. These functions, therefore, are modeled and incorporated into performance assessment. Radionuclide migration in the engineered barrier is considered to proceed as shown in Fig. 7.4.

1. After waste form emplacement, groundwater begins to infiltrate the bentonite buffer, and the groundwater causes general corrosion of the overpack and penetrates it. In this way, after a long period of time after emplacement, the groundwater reaches the vitrified waste forms. At this time, the overpack is deemed to have lost its confinement function.
2. The infiltrating groundwater comes into contact with the vitrified waste forms, and the dissolution and alteration of the vitrified waste forms proceed at a certain rate. The radionuclides are leached out of the vitrified waste forms into the groundwater according to the relative contents of different radionuclides in the forms.
3. Chemical species that are soluble in water remain dissolved in the groundwater. However, those less soluble than the glass component such as hydroxides precipitate on the surfaces of waste forms in the form of their respective stable compounds. The concentration of a radionuclide in the vicinity of vitrified waste forms or in the buffer material is limited by the solubility of each chemical species (not the nuclide). This is called solubility-limited dissolution. Since the deep underground is in an oxygen-poor, reducing environment, many metals are nearly insoluble. In the presence of isotopes, the solubility of the element is assumed to be distributed between the isotopes.

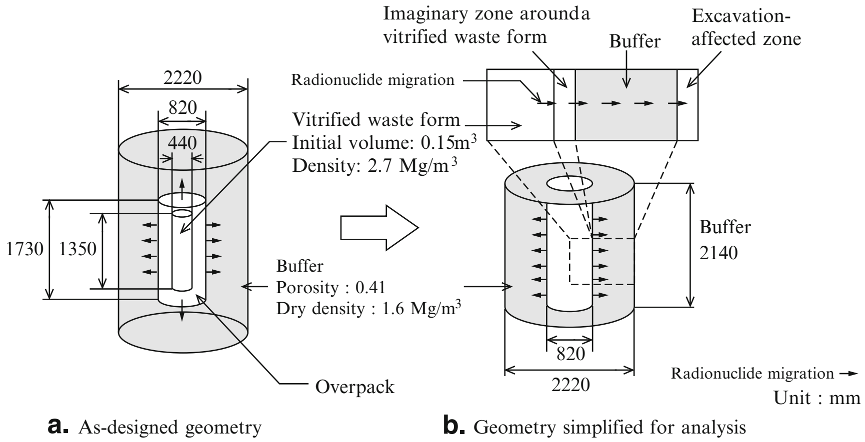


Fig. 7.5 Model for analysis of radionuclide migration in engineered barrier

4. The radionuclides leached out of the vitrified waste forms are transported through the corroded overpack and subsequently through the bentonite buffer by diffusion while being adsorbed during that process.
5. Radionuclides that have reached the outer surface of the buffer migrate to the host rock functioning as a natural barrier. Although the host rock adjoining the repository has lost its original properties under the influence of the excavation during repository construction, this effect is ignored here.

Vitrified waste forms and the bentonite buffer surrounding them are cylindrical in shape as shown in the left half of Fig. 7.5. For mathematical simplicity, this geometry is simplified as shown in the right half of the same figure. In the analysis, the rate of radionuclide release from vitrified waste forms and then the transport of leached radionuclides through the buffer are taken into consideration, and, finally, the source term, i.e., the rate of radionuclide release from the buffer to the surrounding host rock is calculated.

When solubility-limit mechanism as described in step (3) above is implemented in the model, it is necessary to consider the effects of coexisting isotopes of the same element, which share the elemental solubility. The concentration of isotope j of element i, C_{ij}, is in the water phase in the vicinity of the precipitate of element i is approximated by assuming that the solubility C_i* of the element is prorated by the abundance ratio of isotope j in the waste form:

$$C_{ij} = C_i^* \frac{A_{ij}}{\sum_j A_{ij}} \tag{7.6}$$

where A_{ij} is the mass of isotope j of element i.

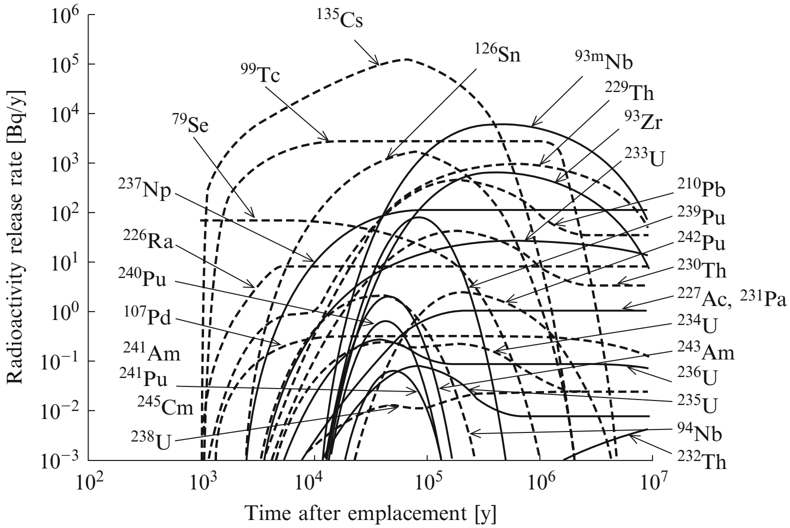


Fig. 7.6 Source term from engineered barrier to surrounding host rock in terms of the rate of radioactivity released

Figure 7.6 shows an example of a source term obtained for a vitrified waste form. The flux (radioactivity release rate [Bq/y]) from the engineered barrier to the surrounding host rock is shown as a function of the time after emplacement. In this figure, radionuclide release from the waste form is assumed to start 1,000 years after emplacement when infiltrating groundwater has reached the surface of the waste form after corrosion of the overpack. It is observed that all curves rise at that time.

In the figure, it is observed that the release rate of ^{135}Cs at the outer surface of the buffer is the greatest among other radionuclides. Because Cs is highly soluble, it is released congruently with the waste form dissolution without being limited by its solubility. Furthermore, the mass contained in the waste form is relatively large, compared with other radionuclides. Due to weak sorption of Cs to the buffer, this isotope will soon appear at the outer surface of the buffer 1,000 years later. At the dissolution rate of the vitrified waste forms used for the analysis, the waste forms will completely dissolve in 70,000 years after the overpack is damaged. Consequently, after peaking at 70,000 years, the release rate of Cs decreases.

After several hundred thousand years, ^{93m}Nb becomes dominant. The half life of ^{93m}Nb is as short as about 16 years. Because ^{93m}Nb is in secular equilibrium with the parent nuclide ^{93}Zr (half life: 1,530,000 y), the curves for the two nuclides are parallel. Since the sorption distribution coefficient of ^{93m}Nb is smaller than that of the parent nuclide, the release rate of ^{93m}Nb is greater than that of the parent nuclide.

Many actinides show relatively low release rates. The reason for this is that most of the radionuclides in this category precipitate after they are released by waste-form dissolution because of their low solubility. Precipitated radionuclides gradually dissolve into groundwater over a long period of time, and migration occurs slowly because of their high sorption tendency.

As these observations indicate, the source term is characterized by the solubility of radionuclides and the sorption property of the buffer.

7.2.3 Migration of Radionuclides in Soil and Rock

Radionuclides that have been released from the repository migrate along groundwater pathways. Groundwater flow in the real world is complex because it varies depending on geological conditions. Furthermore, with current knowledge, it is not possible to mathematically express the reactions of radionuclides with rocks and minerals accurately with microscopic details. In the performance assessment, modeling is carried out by representing three-dimensional flow with one-dimensional flow; representing groundwater flow velocity, which varies spatially in the real world, with the average flow velocity; or simplifying the migration of radionuclides in groundwater by applying known chemical theories.

The concrete vault disposal in Sect. 7.2.2 is considered as the source term here for simplicity. Seepage water containing dissolved radionuclides leaches out from the bottom of the vault into groundwater and joins the groundwater flowing underground. The concentration of radionuclides in the groundwater downstream of the vault is calculated.

The migration of radionuclides in a porous medium such as soil is expressed with a one-dimensional advection–dispersion equation.

$$R_i \frac{\partial C_i}{\partial t} + u \frac{\partial C_i}{\partial x} - D \frac{\partial^2 C_i}{\partial x^2} + R_i \lambda_i C_i - R_{i-1} \lambda_{i-1} C_{i-1} = 0 \tag{7.7}$$

where

C_i: concentration in groundwater of the ith nuclide in the decay chain [Bq/m^3],
u: groundwater flow velocity [m/s],
$R_i = 1 + \rho_b K_{d,i}/\theta$: retardation factor of the ith nuclide,
ρ_b: bulk density of soil [kg/m^3],
θ: porosity of soil, and
$K_{d,i}$: sorption distribution coefficient of the ith nuclide [m^3/kg]

It is assumed that soil is homogeneous, and groundwater flow velocity u, soil porosity θ and the sorption distribution coefficient K_d are constant. It is also assumed that the retardation factor R is constant and remains unchanged. The last term $(i - 1)$ on the left hand side expresses the contribution of the parent nuclide.

In Eq. (7.7), D [m^2/s] is the dispersion coefficient. In an advection–dispersion field, all substances are subjected to two types of action: diffusion due to the Brownian motion of molecules, that is, molecular diffusion of substances dependent on the concentration gradient, and dispersion due to the disturbance of water in the advection field, that is, the heterogeneity of the medium's flow velocity. For a porous medium like soil, as shown in Eq. (8.15) of Chap. 8, the dispersion coefficient can be described by the product of groundwater flow velocity and a constant called dispersion length, which expresses the characteristics of the medium. When advection increases and exceeds a certain limit, dispersion becomes dominant over molecular diffusion due to the Brownian motion. In the advection–dispersion equation, the dispersion coefficient is given as the sum of the diffusion coefficient and the dispersion coefficient.

In the equation shown above, t [s] is time, which is taken to be 0 when the radionuclide begins to migrate in groundwater, and x [m] is the distance. The vault size is not taken into consideration here; the vault is regarded as a point source, and its location is taken to be $x = 0$. It is assumed that at $t = 0$, the soil did not contain any radionuclide. Hence, the initial condition is $C(x, 0) = 0$ $(0 < x < \infty)$ for each nuclide in the decay chain. There are two boundary conditions. One is the concentration at $x = 0$. For the first nuclide in the decay chain, or a nuclide that does not constitute a part of the decay chain, the first condition is given, by using the source term Eq. (7.5) derived in the preceding section, as Eq. (7.8).

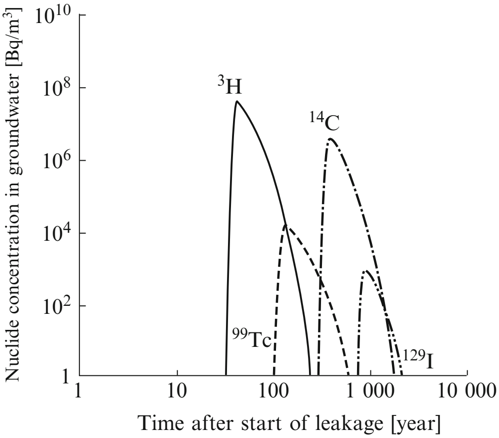
$$C(0,t) = \frac{C_0}{Q}\exp\left[-\left(\lambda + \frac{v}{L\theta R}\right)t\right] \tag{7.8}$$

In the above equation, Q is the dilution ratio for the leachate from the vault diluted by groundwater. For a nuclide in the decay chain, a different formula from Eq. (7.8) is used. Chap. 9 "Solving Diffusion Equations and Advection–Dispersion Equations for Radioactive Materials" shows solutions dealing with decay chains.

The other boundary condition is $C(\infty, t) = 0$. This is obviously an approximation, and this condition assumes that the concentration at a point fairly distant from $x = 0$ is negligibly small compared with the concentration at $x = 0$. By using this approximation, an analytical solution to Eq. (7.7) can be obtained. An analytical solution in the case where a decay chain is not taken into consideration is applied in Exercise 1 of this chapter. Figure 7.7 is an example of the calculation made by using that solution and shows changes in radionuclide concentration at a point 50 m downstream from the vault. Since K_d varies from nuclide to nuclide, the migration rate in groundwater varies, and there are differences in the time of arrival. This indicates that sorption phenomena expressed by K_d play an important role in ensuring safety in radioactive waste disposal.

As an example, assume that 1 g of soil is made to come into contact with 1 liter of water solution of a certain radionuclide. Many K_d values are obtained from this kind of experiment. If 10 % of the radionuclide in water is adsorbed onto soil particles, K_d in this case is 0.111 m^3/kg. If the porosity and bulk density values shown in Fig. 7.7 are used, a retardation factor R of 445 is obtained. This means that this radionuclide moves at only 1/445 of the velocity of groundwater. Arriving at the biosphere from the repository takes a very long time, and the radioactivity decreases during that period because of radioactive decay. The retardation effect by

Fig. 7.7 Radionuclide concentration in groundwater at a point 50 m downstream from the vault. $\rho_b = 1{,}600\ \mathrm{kg/m^3}$, $\theta = 0.4$, $D = 0.04\ \mathrm{m^2/y}$, $u = 10\ \mathrm{m/y}$, $K_d = K_{d,s}$. For source term-related numerical values, see Exercise 1 at the end of this chapter



sorption occurs in the form of the isolation of the radionuclide in the ground. The fact that various barrier materials, rock and soil at disposal facilities can sorb radionuclide effectively is scientific basis for that the underground disposal of radioactive waste is a method for effectively reducing the amount of radioactive materials to be released to the biosphere.

This is true not only in the performance assessment of a shallow underground disposal system but also in the deep geological disposal of high-level radioactive wastes. Since the velocity of groundwater flow is lower at a greater depth than at a shallow level, it can be inferred that the transport of materials at great depths is very slow. When modeling radionuclide transport at great depths, however, it is necessary to take into account a possibility that the host rock may be characterized as porous media such as soil or as fractured media. When the rock is characterized as a fractured medium, matrix diffusion needs to be incorporated in the radionuclide transport model, which does not need to be dealt with when considering a porous medium model.

Figure 7.8 illustrates a groundwater scenario model associated with geological disposal. The quantity of radioactive material that migrates from the repository buffer to the rock, that is, the source term determined in Sect. 7.2.2 is the input in the analysis of radionuclide transport in geological formations. Radionuclides that have migrated from the repository first move through a fracture in the rock (the "100 m" region in the figure) and enter the fracture fault zone. In Japan, the possibility of existence of fault fracture zones cannot be denied. It is therefore assumed that they are major pathways that transport radionuclides from great depths to the surface biosphere. Thus, we need to analyze radionuclide transport through fracture and fault zones.

As discussed in Sect. 7.2.1, a typical model used to deal with a fractured medium in the performance assessment is the parallel plate model. The concept of the model is illustrated in Fig. 7.9. It is assumed that the fracture is the only groundwater pathway, and groundwater is stationary elsewhere. However, rocks, which are

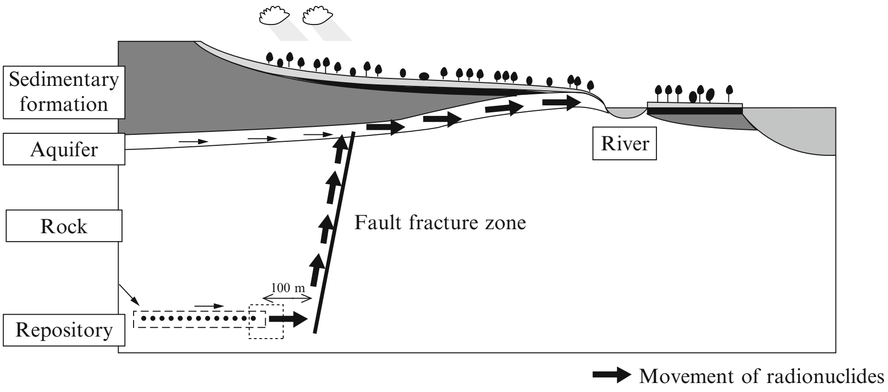
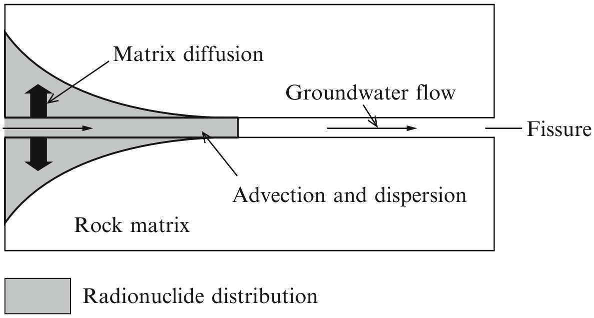


Fig. 7.8 Groundwater migration scenario model (Adapted from Ref. [14])

Fig. 7.9 Mass transport model with planar parallel fractures



aggregates of minerals, may be regarded as porous media with pores formed along mineral grain boundaries. This means that radionuclides can move in the direction perpendicular to a fracture. The driving force in this case is diffusion. This kind of movement to the matrix of rock is called matrix diffusion. In cases where the groundwater-mediated movement of materials is very slow as at great depths, the movement of materials driven by diffusion cannot be ignored. When matrix diffusion is taken into consideration, radionuclide migration through a fissure fracture can be expressed with the following advection–diffusion equation.

$$R_i \frac{\partial C_i}{\partial t} + u \frac{\partial C_i}{\partial x} - D \frac{\partial^2 C_i}{\partial x^2} + R_i \lambda_i C_i - R_{i-1} \lambda_{i-1} C_{i-1} - \frac{F}{b} D_e^m \frac{\partial C_t^m}{\partial y} \bigg|_{y=0} = 0 \quad (7.9)$$

The last term on the left hand side expresses matrix diffusion. Not all the interfacial area between the fracture and the rock matrix contributes to matrix diffusion. The ratio of the area contributing to matrix diffusion is given as F.

Here, $2b$ [m] is the fracture aperture and y [m] is the coordinate distance from the center of the fissure to the matrix. ${D_e}^m$ [m^2/s] is the effective diffusion coefficient in the matrix, derived by modifying the diffusion coefficient in free water to allow for pore structure. Its value can be obtained experimentally. The retardation factor R is relevant to the sorption to the fracture-filling minerals. In a more simplified analysis, it is sometimes assumed that fractures are open and not filled with minerals. In such cases, $R = 1$.

In the matrix, diffusion is the only driving force for the transport of materials. Radionuclide transport is therefore expressed with the diffusion equation:

$$R_i^m \frac{\partial C_i^m}{\partial t} - D_e^m \frac{\partial^2 C_i^m}{\partial y^2} + R_i^m \lambda_i C_i^m - R_{i-1}^m \lambda_{i-1} C_{i-1}^m = 0 \tag{7.10}$$

where the superscript m means that the indicated item is related to the matrix. The governing Eqs. (7.9) and (7.10) are related by two boundary conditions.

$$C_i(x,t) = C_i^m(x, y = 0, t), \left.\frac{\partial C_i^m}{\partial y}\right|_{y=\infty} = 0$$

The former condition requires the concentration continuity at the interface between the fracture and the rock matrix. The latter condition requires that the radionuclide concentration at infinity in the matrix be 0. To what depth materials can actually move can be estimated through observation of natural phenomena, and it is generally said that it is at most 0.1 m or so. Matrix diffusion results from the difference between the radionuclide concentration in the fracture and the concentration in the matrix. When the concentration in the fracture is higher, therefore, the radionuclide diffuses further into the matrix, and when the concentration in the fracture decreases, it diffuses in the opposite direction toward the fracture. In other words, by matrix diffusion, instead of moving directly toward the biosphere with groundwater, the radionuclide moves in a roundabout way; essentially, it moves so as to lower the peak of radionuclide concentration occurring in the biosphere. During the "roundabout" movement, radionuclide concentration decreases because of radioactive decay.

The radionuclide that has thus moved through the fracture in the host rock enters the fault zone. If it is assumed that in the fault zone, too, the radionuclide migrates by advection and dispersion while undergoing matrix diffusion and sorption, and radionuclide transport can be described by Eq. (7.9). Because data values differ between the fracture and the fault zone due to differences in hydraulic characteristics, appropriate values should be selected according to their respective hydraulic and fissure characteristics.

The groundwater containing the radionuclide that has migrated through the fault zone joins the near-surface aquifer and arrives at the biosphere. When groundwater enters the fault zone from the fracture in the host rock and when it enters the aquifer

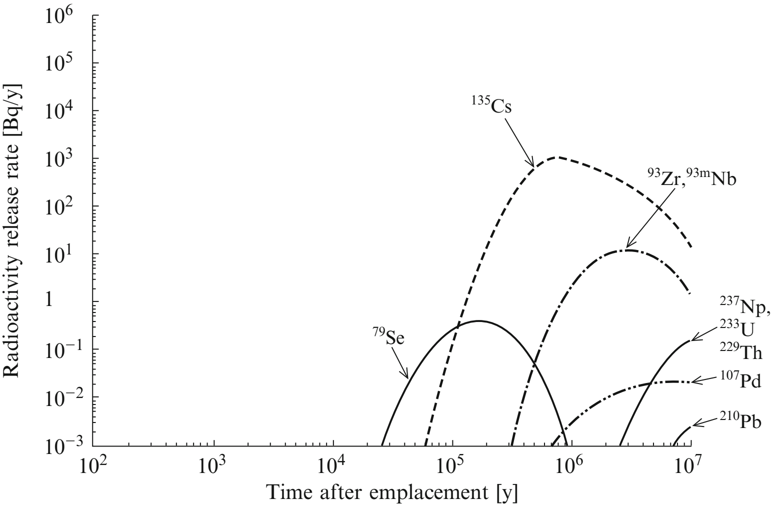


Fig. 7.10 Radioactivity release rate from the fault zone to the biosphere [14]

from the fault zone, dilution occurs and, as a result, the radionuclide concentration decreases abruptly. The radionuclide flowing into the biosphere after undergoing this process is the input to the assessment of radionuclide transport in the biosphere dealt with in the next section. Examples of such calculation are shown in Fig. 7.10.

While the radionuclides confined in waste forms pass through the engineered barrier, migrate through the ground and finally reach the biosphere at the ground surface, they undergo a variety of phenomena. The preceding section and this section have described the transport of radionuclides with a few governing equations on the basis of mass transport theory. It may easily be noted that those equations are results obtained by simplifying and ignoring various phenomena and making conservative assumptions.

Progress is always being made, however, in the field of modeling as a result of advances in the techniques to observe groundwater hydrology and the geological environment; advances in science concerning the interaction between chemical elements, minerals and water at great depths underground; and advances in computer science. The model described above is a very simple one compared with the current knowledge at this writing. In fact, even in the area of groundwater flow velocity, changes and distributions are being taken into consideration and modeling is carried out accordingly, and research is being conducted to perform analyses as realistically as possible by applying the best of the available knowledge.

Now, again the boundary condition Eq. (7.8) for the advection–dispersion equation Eq. (7.7) is considered. Eq. (7.8) specifies the condition at the location where the disposal facility and the geological medium are linked in terms of concentration. The system for analyzing the shallow underground disposal facility considered here, radionuclide leakage from there and the subsequent radionuclide migration, is the same as the system used to calculate clearance levels for radioactive waste described in Chap. 4 (see also Fig. 8.1 of Chap. 8). Clearance levels for this system were determined through the analysis beginning with Eq. (8.12). It should be noted, however, that the mathematical models expressing the boundary conditions are different. In this section, the boundary condition is given in terms of concentration, while in the analysis described in Chap. 8, the boundary condition is given in terms of flux as shown in Eq. (8.12). It is thought likely that Eq. (7.8) is the result obtained by making extremely conservative assumptions about the barrier performance of the disposal facility as can be seen from the system to which the equation was applied (Fig. 7.3).

Chapter 9 "Solving Diffusion Equations and Advection–Dispersion Equations for Radioactive Materials" describes how to solve equations under some typical conditions. Although many of the advection–dispersion equations used in connection with radioactive waste disposal, including those involving matrix diffusion, require numerical solution methods, the equations described in Chap. 9 are ones that can be solved analytically. They should be reviewed for self-study.

7.2.4 Radionuclide Transfer in the Biosphere

The analysis of radionuclide transfer in the biosphere is to be made in the performance assessment for the purpose of presenting assessment results in the form of indices for the system performance in terms of influence of radiation from radionuclides. After reaching the biosphere in which humans live, groundwater containing radionuclides could be taken in by humans in food so as to cause internal exposure or enters soil so as to cause external exposure. The evaluation of such influence is termed radiation exposure evaluation. Radiation exposure evaluation begins with tracing radionuclide transfer pathways.

The first thing to keep in mind in the biosphere pathway analysis is that it is not possible nor attempted to predict changes in the biosphere in the distant future. In the performance assessment of geological disposal, the time horizon for assessment is by far longer than the life span of humans. Nevertheless, it is not completely impossible to describe with a certain level of confidence future geological changes based on past events. As can be seen, however, from the example of the Sahara Desert, which was transformed from a dense forest region to a desert in only thousands of years, ground surface changes are fast in the geological disposal performance assessment period, and it is by no means possible to predict changes in the condition of humans living there. Furthermore, the purpose of showing the performance of the disposal system does not even try to describe in detail the conditions in the biosphere in the distant future. What is done, therefore, is to construct models with certain abstraction; this is called "stylization." Stylization does not aim to depict the distant future realistically. Instead, it is an approach for quantifying radiological impacts of geological disposal on future generations of humans in the context and framework of the present regulation and safety concepts.

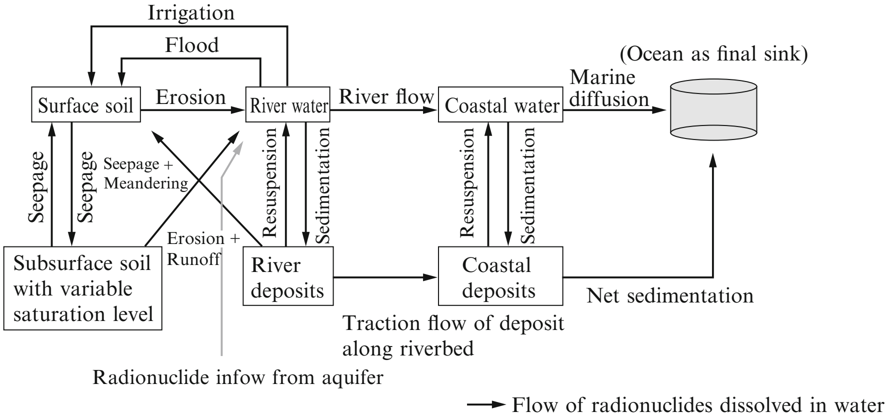


Fig. 7.11 Model of radionuclide transfer in the biosphere [19]

The important advantage of this stylization is that this procedure makes quantitative discussion possible.

The assessment of the influence of radionuclides in the biosphere is considered in two stages: the radionuclide transfer pathways in the biosphere and the possible radiation exposure along pathways.

As shown in Fig. 7.8, radionuclides that have migrated at great depths pass through the fault zone and then the aquifer, and to the biosphere. Entrances to the biosphere include rivers, wells and seas. Figure 7.11 shows how the radionuclides that have flown into a river from an aquifer move into the biosphere. As shown, the radionuclides that have flown into the river move between various components of the living environment, such as riverbed deposits and surface soil, are transported by river flow to the coastal water, and eventually dissipate into the ocean.

If the evaluation time scale is long as in the evaluation of material movement in the environment and the rate of material transfer is high (i.e., fast), the concept called the box model is often used. In individual compartments (boxes) such as “river water” and “coastal water” in Fig. 7.11, materials are mixed well and a state of equilibrium is maintained. Material transfer shown with an arrow such as “sedimentation” and “resuspension” is dealt with kinetically. In this model, therefore, kinetic data expressing material transfer between boxes are needed.

Like pathways for clearance level calculation (Figs. 4.1 and 4.2), possible human exposure pathways are assumed from every situation in which humans can come into contact with radioactive materials (Fig. 7.12). For each of the radionuclides under consideration, exposure doses along the assumed pathways are calculated by using a variety of data such as data on the migration from soils, deposits, river waters, coastal waters, etc., concentration in the food chain, the amount of human food intake, time of contact with rivers and soils and the exposure dose conversion factor. Exposure doses from the intake of agricultural products are relatively large. Consequently, among the three occupational groups shown in Fig. 7.12, agricultural workers are more seriously affected than the other two groups.

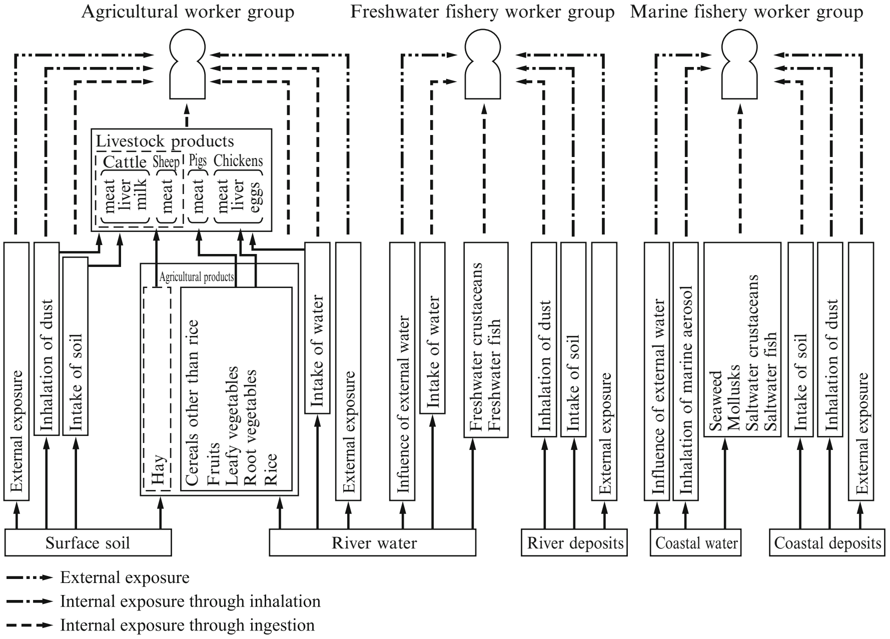


Fig. 7.12 Pathways through which representative groups are affected by radionuclides [19]

Figure 7.13 shows annual exposure doses calculated as a function of time after emplacement for the groundwater scenario in which radionuclides leach out of vitrified waste forms emplaced in the geological repository, pass through an engineered barrier, move in groundwater, and finally emerge in the biosphere. According to these results, the exposure dose reaches the maximum value of 0.005 μSv/y about 800,000 years after emplacement.

The radionuclide that governs the annual exposure dose varies depending on the time after emplacement. The governing radionuclide for the first 200,000 years or so is ^{79}Se, followed by ^{135}Cs until 6 million years after emplacement, which is then followed by ^{229}Th until several tens of million years after emplacement. The radionuclide that causes the maximum annual dose is ^{135}Cs. Over a very long period of time like the time scale shown by the horizontal axis in Fig. 7.13, many generations of humans will come and go. The resultant changes in life styles are not reflected in the mathematical model. This time axis needs to be understood on the basis of the concept of stylization.

Japan does not have established regulatory guidelines for annual doses for geological disposal. Figure 7.13 shows the natural radiation levels in Japan and the safety standard values in other countries mentioned in Sect. 7.1.1. The regulatory body of Japan has stipulated that in the safety assessment of geological disposal, it is a basic consensus to confirm that possible exposure doses imposed on the general public from radioactive materials do not exceed the specified radiation protection levels under appropriate scenarios [20]. The performance

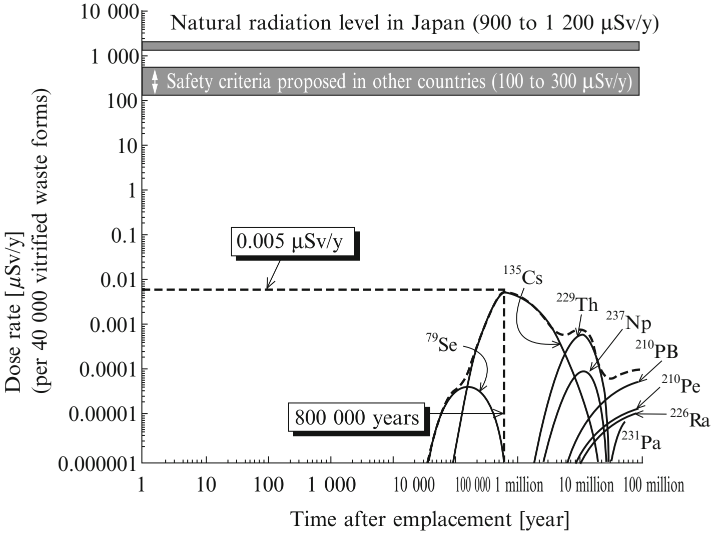


Fig. 7.13 Exposure doses due to 40,000 vitrified waste forms in geological disposal [3]

assessment results obtained from radionuclide migration analyses are one of the bases for showing the safety of geological disposal by comparison with specified safety criteria as shown in Fig. 7.13. When interpreting and using those results, however, it is necessary to conduct careful studies as described in the next section.

7.3 Interpretation and Use of Performance Assessment Results

Performance assessment results provide information that can be used to judge, by making comparison with the safety criteria specified by the regulatory authorities, whether the system under consideration is safe over a long period of time. At stages where a disposal site has not yet been determined, performance assessment results provide information that can be used to judge whether the system concept and design under consideration are appropriate, and help make a decision on the direction of the subsequent stages of geological disposal system development.

7.3.1 Safety

The evaluation of the level of safety of the disposal system by comparison with safety criteria is one of the most important purposes of performance assessment. It is therefore necessary to show performance assessment results in a form that makes comparison with the safety criteria possible. If different safety criteria have been specified for two or more scenarios, it is necessary to show results for each set of those criteria.

In Europe, geological disposal systems had been developed over the years without regulatory authorities' expressly indicating safety criteria. It is not possible to directly verify geological disposal system performance because of a very long evaluation period. There is no choice, therefore, but to use methods of indirect demonstration and verification, and, unlike other engineered structures, is necessary to verify their safety before the systems are in existence. The number of disposal systems that will come into operation in the world by the end of this century will be, at most, ten or so. When evaluating the level of safety of an engineered structure, it is common practice to do so statistically on the basis of data on many accidents. In the case of the disposal system, however, this type of standard approach in safety engineering cannot be used. Furthermore, society has not been ready to deal with the new concept of safety over a period longer than 10,000 years. Thus, it was thought that the determination of safety criteria itself should be based on performance assessment results. And therefore, the engineering development of a disposal system and the institution of safety criteria should proceed concurrently as a concerted effort.

In Europe, the International Commission on Radiological Protection (ICRP) dose limit of 1 mSv/y was first introduced into the domestic laws and regulations of the countries. Then, different dose limits were specified in different countries, namely, 0.25 mSv/y in France, 0.3 mSv/y in Germany, and 0.1 mSv/y in Sweden and Finland. Later, since ICRP recommended dose constraints of 1 mSv/y or less, and less than 0.3 mSv/y, explanations consistent with these values have been made. In Japan, too, deliberations are underway to set reference dose values consistent with the ICRP criteria. Concurrently, the design of disposal facilities and research and development necessary for that purpose are being conducted.

In the U.S., safety criteria were established as early as the 1980s, when disposal sites had not been determined, on the rationale that judgment cannot be made if there are no criteria. Later, safety criteria specific to the Yucca Mountain repository were determined, but the (generic) safety criteria determined in the 1980s were applied to the WIPP. Not only has the Department of Energy carried out performance assessments repeatedly, but also the Nuclear Regulatory Commission conducted performance assessments on its own partly for the purpose of enhancing its ability to conduct safety examination. Thus, in the U.S., performance assessments were carried out by both waste disposers and safety assessment authorities. The risk-informed, performance-based regulation approach has been adopted in all aspects of nuclear regulation; and under the direction of the Congress, the safety criteria and regulations were reviewed by the National Academy of Sciences. As a result, with respect to the safety criteria adopted for the Yucca Mountain repository, the previously adopted criteria for individual barriers were abolished, on the rationale that the goal can be achieved if the upper limit of the exposure dose rate for disposal is met by the system as a whole.

7.3.2 Technology Development

The disposal project involves stages of site selection, geological environment investigation, engineering barrier performance experiments and the like, which takes as long as several tens of years. In order to carry out R&D over such a long period of time systematically and efficiently, it is good practice to identify, and focus R&D efforts on, FEPs that have important effects on system performance. Performance assessment models and codes can be used to carry out sensitivity analyses to determine R&D priorities. It is also possible through uncertainty analysis to determine which part of the system or which parameter is causing the uncertainty of system assessment results.

In system development, actions taken after main factors affecting system behavior are identified are important. Mainly two types of actions can be taken. One is to reconsider the system itself so that factors that have an important (negative) impact are eliminated. The other is to aim at deepening scientific understanding of those factors. In this section, the former is called an engineering approach, and the latter a scientific approach.

An example of an engineering approach is as follows. Research has shown that the solubility of a radionuclide in groundwater is a parameter that has a dominant impact on the exposure dose rate to be determined finally as a performance indicator. Considerable research has focused, therefore, on the measurement of the solubility of low-solubility elements, particularly actinides, and it has been found that the solubility of actinides is affected by the environment in the engineered barrier. However, although epistemic uncertainty can be reduced by obtaining more accurate understanding on the mechanism for determining the solubility of actinides under certain conditions, environmental changes over time have aleatory uncertainty. This has shown that a relatively high degree of uncertainty is inevitable if the approach to ensure the safety of a disposal system dependent on low solubility of actinides is to be taken. This became one of the factors that led to the initiation of the study on options (e.g., partitioning and transmutation) that can be adopted to reduce the inventory of actinides in disposal systems and change the properties of waste forms. Whether such options are used in combination is determined taking into consideration the cost of the entire system, performance requirements, and the degree of uncertainty.

An example of a scientific approach is as follows. The dissolution of vitrified waste forms is an important phenomenon that determines the source term for geological disposal, and vigorous research efforts were once being made in many countries. A large amount of leaching rate data were obtained and used for performance assessment in each country. It was thought then that even if vitrified waste form leaching rates obtained as part of the results of sensitivity analyses conducted in performance assessment are inaccurate by two or three orders of magnitude because of uncertainty, the calculated values of exposure doses that will peak several hundred thousand years later are hardly affected by the uncertainty of the calculated leaching rates of vitrified waste forms because such uncertainty is overwhelmed mainly by the uncertainty of the solubility of radionuclides. The results of subsequent research, however, indicated that the solubility of radionuclides, particularly actinides, is greatly affected by their chemical environment. It is highly likely, therefore, that poor understanding of changes in the chemical environment caused by the dissolution of vitrified waste forms is the main cause of the uncertainty of the solubility of radionuclides. It can be said that the objective of the scientific approach is to reduce epistemic uncertainty as much as possible by gaining a deeper understanding. As in the engineering approach, the degree of understanding to be pursued (from the viewpoint of the achievement of disposal, aside from the scientific interest of scientists) is to be determined in view of the total cost of the system, performance requirements and the degree of uncertainty. This must not be used, however, as a reason for abandoning scientific pursuits.

These two approaches are not exclusive; they should be pursued concurrently. As technology development proceeds and understanding becomes deeper, it is important to improve the applicable models. For example, usually, previously developed models should not be used without making modifications when the engineered barrier configuration is changed, a new site is considered, options for changing the properties of wastes such as partitioning and transmutation are used in combination or a new discovery has been made of a mechanism. It is necessary to study the coupling of FEPs and changes in environmental conditions in detail and evaluate the validity of the models under consideration. Since many of the conventional models are based on various conservative assumptions, it is necessary to evaluate their validity under new conditions and develop new models if necessary.

7.3.3 Public Consensus Building Process and Policy Decision Making

Policy decision making is needed at each stage of a geological disposal project. At the initial stage, the decision is made whether or not to proceed with geological disposal. For a country making use of nuclear power on a large scale like Japan, the decision at this stage is self-evident, and the decision not to carry out geological disposal is an unlikely choice. For a country utilizing nuclear power on a smaller scale by operating only a small number of power reactors, however, options within an international framework such as international disposal facilities are among important considerations. Even for such countries, for example, the performance assessment R&D program that has been carried out by Switzerland will be very informative. In fact, experience accumulated in connection with Switzerland's disposal program is greatly affecting the European Union's ongoing international repository study program. If a country has decided to carry out geological disposal in the country, it is necessary to evaluate the feasibility of geological disposal. In the case of Japan, it is generally thought that feasibility was verified in the “H12 Report” (2000 Report) [3]. There are also countries such as France that made a policy decision at the initial stage, taking into consideration partitioning and transmutation as well as geological disposal. There are also countries that are considering alternative options such as the U.S., which had been working on development for direct disposal of spent fuel. Thus, policy decision making often takes time because it necessarily involves other related decisions such as what to dispose of and how to dispose it. Even when a decision has been made, it may be reconsidered later depending on the situation.

After making a decision to carry out geological disposal, the most important policy decision is about site selection. In the area of site selection, trial-and-error efforts have been continued in each country, but there is as yet no commonly recognized methodology or solution. In the U.S., there are two examples, namely, WIPP and the Yucca Mountain project, the former of which is thought to be a success and the latter is, at least at present, thought to be a failure. In Canada, detailed engineering studies on the concept of the direct disposal of spent fuel in stable granite formations had been completed by the 1990s, but the scheme was practically abandoned because public consensus was not reached. Under these circumstances, the successes of Sweden and Finland in public consensus building and site selection are very informative. In Canada, too, efforts have been made since the 2000s to find a way to reach public consensus, and a comprehensive agreement has been successfully reached on matters including geological disposal.

Thus, as a general trend, more attention is being paid to the bottom-up approach, which aims to determine a framework through public discussions on how to solve radioactive waste disposal problems or what constitutes a success in solving those problems, rather than the top-down approach, which aims to explain the central government’s decision to the local residents in the disposal site area and make it understood. Public discussions can be done in various ways depending on the conditions in each country, but the important thing is to provide information needed in the discussion process in a clear and easy-to-understand way.

Performance assessment, therefore, is very important, and its results need to be interpreted and used carefully. Performance assessment provides information that is most important for policy decision making, that is, information on safety. In many cases, performance assessment results indicate that the disposal system is sufficiently safe. According to the conventional concept of performance assessment, the goal of performance assessment has now been achieved—nothing more, nothing less. How those results are used is left to policy executors and the public. In reality, however, it has been shown in advanced countries such as the U.S. and Canada that disposal cannot be carried out simply by showing that performance assessment results indicate a sufficient level of safety. Some say that this is not a matter of performance assessment but a matter of the usage of performance assessment results. Examination, however, of how performance assessment results are used for public consensus building and policy decision making reveals that there are cases where problems lie in performance assessment itself. Three points are pointed out below.

The first point concerns performance indices. If the exposure dose rate after disposal is used as an index, it is necessary to model the biosphere as the final stage of the nuclide migration process. As mentioned in Sect. 7.2.4, it is impossible and pointless to predict human life styles in the distant future. In performance assessment, stylization is made by using the present conditions such as population distribution and diet as the basis for the development of a biosphere model. Performance assessment results, therefore, should be regarded not as estimates of human exposure doses in future but as an index by which to judge whether estimated amounts of nuclides released into present day society are permissible. For this reason, in performance assessment, the terms "estimation," "prediction" and "simulation" should be carefully avoided.

The second point is this. As mentioned earlier, performance assessment is carried out to evaluate the level of safety of the disposal system. In the coming years, however, there will be a growing need to compare two or more sites, system concepts, designs and technology options. It may also become necessary to optimize the disposal system, for example by increasing disposal capacity or minimizing cost, even at an advanced stage of the program. There will be growing demand for performance assessment results and methods to be used for such comparison or optimization. Performance assessment, however, will be useless in communication with the public and policy decision making if such needs and demands cannot be met. A number of attempts have already been made to make such comparisons or optimizations by using existing performance assessment models. An example is the study on the influence of partitioning and transmutation on geological disposal introduced in Chap. 2. In the U.S., before the Yucca Mountain site was selected, a detailed comparative study of five and later three candidate sites was conducted. However, looking back today, when R&D in relevant fields has reached a certain level both in depth and coverage, the study results are lacking in many ways if they are to be used for comparative or optimization research. In particular, further study using detailed models is needed to determine whether or not the conservative assumptions incorporated in many parts of the performance assessment models are applicable to the other sites, systems, designs and technology options to be compared.

The third point concerns the time scale and the time axis. This is closely related to the index-related problem described above as the first point. Although this has already been mentioned earlier in this chapter, the problem is summarized here. The aim of performance assessment is not prediction. When observing and evaluating system behavior however, the scheme of time cannot be ignored. This is because safety is inextricably linked to system deterioration, and deterioration occurs with time. Because geological disposal includes long-lived radionuclides in the waste, the time scale to be considered, therefore, is far longer than the time scale (several tens of years) of normal human activities, and the evaluation of performance indices must cover a very long period of time (several million years). When thought of in this way, it can be noted that there are different time axes. One of them is the time axis for the progress of deterioration of engineered barriers. This can be predicted and can be verified indirectly by using the scientific approach described in the preceding section. It is possible to reduce epistemic uncertainty to some degree, and the behavior of an engineered barrier can be explained chronologically and scientifically. When a quantity related to the behavior of an engineered barrier is expressed as a function of time (for example, Fig. 7.6), the time measured on the horizontal axis may be thought of as actual time that is not stylized. It may be that phenomena involving natural barriers may be included in this category, but dealing with them separately may be a better way because the degree of aleatory uncertainty can be considerably high. For example, the time measured on the horizontal axis of Fig. 7.10, too, may be thought of as non-stylized actual time. In contrast, the time axis shown in Fig. 7.13 is stylized. Thus, it must be understood that the meaning of this horizontal axis (time axis) is basically different from that of the horizontal axes shown in the two figures mentioned above.

An important point to pay attention to when considering the relationship of performance assessment results with public consensus building and policy decision making is whether or not these complex conditions and performance assessment results that require interpretation can play their expected roles in the public consensus building process. In fact, this difficulty has already been recognized, and one approach currently in use as a means of meeting this requirement is the introduction of the BAT concept mentioned in Chap. 6.

Exercises

1. Draw a graph of changes over time in the concentration C_s [Bq/m^3] of each radionuclide in water in a vault over the period from 1 to 1,000 years after emplacement by using the distribution coefficient source term model (Eq. (7.5)) for concrete vault disposal . The disposal facility conditions and the initial values for the radionuclides are shown in the tables below.

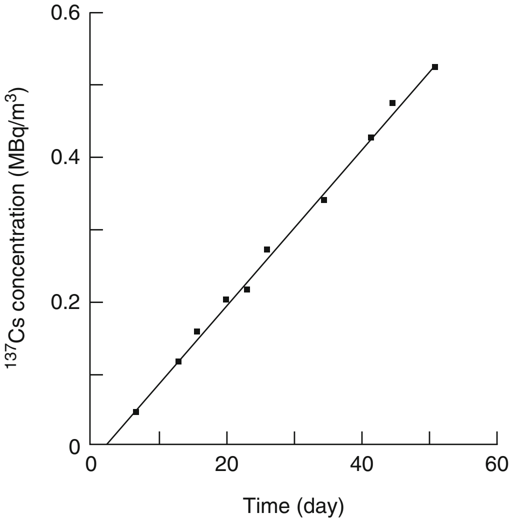
Seepage rate v	0.1 m/y
Vault height L	5.0 m
Porosity in the vault θ_s	0.5
Bulk density in the vault $\rho_{b,s}$	500 kg/m^3

Radionuclide	Half life [year]	Inventory C_0 [Bq/m^3]	Sorption distribution coefficient $K_{d,s}$ [m^3/kg]
^{3}H	12.35	1.0×10^{11}	0
^{14}C	5,730	1.0×10^{10}	0.002
^{60}Co	5.3	1.0×10^{11}	0.01
^{59}Ni	75,000	1.0×10^{7}	0.009
^{63}Ni	100	1.0×10^{12}	0.009
^{79}Se	65,000	1.0×10^{7}	0.005
^{90}Sr	28.8	1.0×10^{12}	0.008

(continued)

Radionuclide	Half life [year]	Inventory C_0 [Bq/m^3]	Sorption distribution coefficient $K_{d,s}$ [m^3/kg]
^{99}Tc	2.13×10^5	1.0×10^7	0.0005
^{106}Ru	1.08	1.0×10^{10}	0.1
^{129}I	1.57×10^7	1.0×10^7	0.005
^{134}Cs	2.06	1.0×10^7	0.1
^{137}Cs	30.2	1.0×10^{12}	0.1

Fig. 7.14 Changes over time in ^{137}Cs concentration in the low-concentration cell



2. A cesium diffusion experiment was conducted by using the through-diffusion cell apparatus (Fig. 9.6) shown in Sect. 9.11 "Diffusion through a Medium of Finite Length." The measurement results obtained in the low-concentration cell are shown in Fig. 7.14. As shown, after a certain period of time, a straight line with a slope of 0.011 MBq/(m^3·day) and an x-intercept of 2.5 days was observed. The Cs concentration on the high concentration side gradually decreased from the initial value (10 MBq/m^3). From these measurement results, calculate the diffusion coefficient and the sorption distribution coefficient. As shown in Fig. 9.6, since the surfaces in contact with the bentonite–sand mixture solution are 20 mm in diameter, diffusion area A is 3.14×10^{-4} m^2; the volume V of water in the cell is 1.1×10^{-4} m^3; the thickness L of the specimen is 12 mm; the porosity ε is 0.4; and density ρ is 1.6×10^3 kg/m^3.
3. On the basis of the description in Sect. 7.2, list examples of aleatory uncertainty and epistemic uncertainty and consider how they affect performance assessment results.

References

1. International Atomic Energy Agency, *Geological Disposal of Radioactive Waste*, IAEA Safety Requirements No. WS-R-4 (Jointly sponsored by the IAEA and the OECD/NEA, 2006)
2. Power Reactor and Nuclear Fuel Development Corporation, *Research and Development on the Geological Disposal of High-Level Radioactive Waste (Fiscal 1991)*, (1992)
3. Japan Nuclear Cycle Development Institute, *Technical Reliability of the Geological Disposal of High-Level Radioactive Waste in Japan: Second Progress Report on Research and Development for Geological Disposal*, 4 vols. (1999)
4. Nuclear Energy Agency of the Organisation for Economic Co-operation and Development, *Scenario Development Methods and Practice: An Evaluation Based on the NEA Workshop on Scenario Development*, Madrid, Spain, May 1999 (2001)
5. Nuclear Energy Agency of the Organization for Economic Cooperation and Development, *Features, Events, and Processes (FEPs) for Geological disposal of Radioactive Waste: An International Database* (2000)
6. Statens Kärnkraftinspektion [Swedish Nuclear Power Inspectorate: SKI], *SKI-Site-94: Deep Repository Performance Assessment Project*, vol 1, SKI Report 96: 36 (1996)
7. J. Ahn, D. Kawasaki, P.L. Chambré, Relationship among performance of geologic repositories, canister-array configuration, and radionuclide mass in waste. Nucl. Technol. **126**, 94–112 (2002)
8. U.S. Department of Energy, *Viability assessment of a repository at Yucca Mountain, Vol. 3: Total system performance assessment*, DOE/RW-0508, 1998
9. N. Chapman, C. McCombie, Principles and standards for the disposal of long-lived radioactive wastes, Waste Management Series, vol. 3, (Elsevier, 2003)
10. J.C. Helton, M.G. Marietta, The 1996 performance assessment for the waste isolation pilot plant, Reliab. Eng. Syst. Saf. (Special Issue), **69**(1–3) (2000)
11. K. Spitz, J. Moreno, *Groundwater Environment Modeling for Engineers*, trans. by the Okayama Ground Water Research Group, Hakuhodo (2003)
12. Y. Ishida, T. Moritomo, Y. Sakaki, Geological environment study for feasibility assessment of subsurface disposal. J. Nucl. Fuel Cycle Environ. **13**, 37–48 (2006)
13. R. Sakai, M. Munakata, M. Ohoka, H. Kameya, *Study on Uncertainty Evaluation Methodology Related to Hydrological Parameters of Regional Groundwater Flow Analysis Model*, JAEA-Research 2009-038 (Japan Atomic Energy Agency, 2009)
14. Japan Nuclear Cycle Development Institute, *Technical Reliability of the Geological Disposal of High-Level Radioactive Waste in Japan: Second Progress Report on Research and Development for Geological Disposal*, vol 3: Safety assessment of geological disposal systems. JNC TN1400 99–23 (1999)
15. T. Iwatsuki, K. Morikawa, S. Hosoya, H. Yoshikawa, Physicochemical parameters (pH, redox potential) of deep groundwater: Measurement and considerations. J. Groundw. Hydrol. **51**, 205 (2009)
16. K. Spahiu, L. Werme, J. Low, U.B. Eklund, *In situ long-term measurements of pH and redox potential during spent fuel leaching under stationary conditions: The method and some preliminary results*, MRS Symposium Proceedings 608, Materials Research Society, 55 (2000)
17. Y. Amano, T. Iwatsuki, S. Ioka, H. Sasao, Microbial effect on redox condition in subsurface environment. J. Nucl. Fuel Cycle Environ. **14**, 61 (2007)
18. Agency for Natural Resources and Energy, Ministry of Economy, Trade and Industry, *Shall we think about geological disposal of high-level radioactive waste?* (2008)
19. Japan Nuclear Cycle Development Institute, *Technical reliability of the geological disposal of high-level radioactive waste in Japan: Second progress report on research and development for geological disposal – Project overview report: Safety assessment of geological disposal systems*. JNC TN1400 99–20 (1999)
20. Nuclear Safety Commission of Japan, *Basic concept of safety regulation for high-level radioactive waste disposal* (first progress report) (2000)

Chapter 8
Appendix A: Derivation of Clearance Levels

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Like the derivation of exhaust air and waste water radioactivity concentration limits based on the reference dose values for radiation protection, clearance levels are derived by following these steps:

1. Determine reference dose values for clearance level derivation.
2. Select evaluation items for clearance.
3. Select evaluation pathways for exposure dose evaluation and persons to be evaluated.
4. Construct exposure dose evaluation models along the evaluation pathways.
5. Set scenario parameters to be used as derivation conditions.
6. Derive radioactivity concentrations of nuclide corresponding to reference dose values.

Each of the steps is described below. This chapter, however, does not cover all clearance level derivation methods. For further details, see the appendices to the clearance report for light water reactor and gas-cooled reactors used for power generation and research reactors [1] and the clearance report for radioisotope (RI)-using facilities and radiation generators [2]. The scenario parameter symbols used in this chapter "Derivation of Clearance Levels" is the same as that used in connection with the discussions on the clearance system. The symbols used, therefore, are common to Chap. 4 "Clearance" but are not necessarily the same as

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The nuclear regulatory system in Japan has been changed significantly after the Fukushima Daiichi Nuclear Power Station accident in March 2011. Descriptions in this chapter have been translated from the book originally published in Japanese before the accident, with minimal update where appropriate. The Nuclear Regulation Authority newly established after the accident has not completed its review for the guidelines and regulations established by the former Nuclear Safety Commission. In this chapter, guidelines set by the NSC have been adopted.

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those used in other chapters of this book. There are also cases where scenario parameters of the same kind are represented by different symbols because many scenario parameters are used. For example, flux may be represented by A or J. The units used are the ones customarily used in this field and, therefore, are not necessarily SI units. Attention should be paid to the symbols and units used for each equation.

8.1 Reference Dose Values

As mentioned in Chap. 4, the reference dose values used to derive clearance levels are an individual effective dose of 10 μSv/y or an equivalent dose at the skin of 50 mSv/y. For each evaluation pathway, the radioactivity concentration of each nuclide corresponding to the chosen reference dose value is derived, and the lowest radioactivity concentration thus obtained is taken as the clearance level for that nuclide.

8.2 Evaluation Items

Evaluation items for the clearance level derivation are concrete and metals generated by the dismantling of reactor facilities and nuclear fuel use facilities. In the case of RI-using facilities or radiation generator, incineration ash is added to the evaluation items mentioned above, and in uranium-handling facilities, only metals are evaluated. Shown below are derivation conditions for evaluation items under which clearance levels are derived for reactor facilities. Derivation conditions for evaluation items for the clearance levels derivation for uranium-handling facilities are shown in the uranium clearance report [3]. Derivation conditions for evaluation items under which to derive clearance levels for RI-using facilities and radiation generators are described in the RI clearance report [2].

It is assumed that the weight of the wastes, W, to be buried is 500,000 tons, which consists of 50,000 tons of cleared items and 450,000 tons of “non-radioactive wastes.” The mixing ratio between the wastes and cleared items is referred to as the weight ratio of the cleared items in the wastes, F_{WC}, and in this case, it is 0.1. In recycling, both the weight ratio of the cleared metals in the metals to be recycled (F_{MC}) shown in Fig. 4.3 and the weight ratio of the cleared concrete in the concrete to be recycled (F_{CC}) shown in Fig. 4.4 are assumed to be 0.1.

8.3 Evaluation Pathways

Evaluation pathways related to recycling and reuse are as shown in Fig. 4.1. Cleared metals are supposed to be recycled as metal products such as consumer goods and construction materials as shown in Fig. 4.3, whereas cleared concrete is supposed to be recycled as construction materials and parking lot base course materials as shown in Fig. 4.4. The evaluation pathways related to burial disposal are shown in Fig. 4.2. It is assumed that cleared concrete and cleared metals are to be buried at a waste disposal facility of the same size as disposal facilities for industrial wastes (length L_D: 200 m, width W_D: 200 m, depth H_D: 10 m).

Persons to be evaluated are adult workers who directly work on or with cleared items, adults engaged in agriculture, adult residents and 1- or 2-year-old child residents. For these types of people, models are made along the evaluation pathways, and exposure doses are derived for four types of exposure, namely, external exposure, inhalation intake exposure, ingestion intake exposure and skin exposure.

8.4 Exposure Dose Evaluation Models

8.4.1 External Exposure Dose Evaluation Model

Let $C_m(i)$ [Bq/g] be the radioactivity concentration of nuclide i contained in cleared items or recycled products. Then, the external exposure dose $D_{EXT}(i)$ [Sv/y] due to cleared items or recycled products such as metal products, construction materials or parking lot base course materials can be derived, as shown in Eq. (4.1) given in Chap. 4, as

$$D_{EXT}(i) = C_m(i) \cdot S \cdot t_f \cdot \frac{1 - \exp(-\lambda_i t_i)}{\lambda_i t_i} \cdot D_{CF,EA}(i) \tag{8.1}$$

where

S: shielding factor for external radiation,
t_f: annual exposure time during the exposure scenario,
λ_i: decay constant of nuclide i [y^{-1}],
t_i: exposure time during the exposure scenario [y], and
$D_{CF,EA}(i)$: dose conversion factor for external exposure to nuclide i [(Sv/y)/(Bq/g)].

For the annual exposure time during the exposure scenario t_f, the ratio of time during which a recycled product is used in a year is taken. The shielding factor S for external radiation is 1 if the person to be evaluated is not shielded by a wall-like barrier during the service life of the recycled product. If exposure continues for a long period of time, it is necessary to take into account a decay in radioactivity concentration during the period of external exposure to nuclide i.

The radioactivity concentration $C_m(i)$ of nuclide i in cleared items or recycled products varies along evaluation pathways and is derived as shown below.

In scrap transportation work and landfill work, on the assumption that the wastes are a mixture of cleared items and “non-radioactive wastes,” $C_m(i)$ is derived as follows:

$$C_m(i) = C_{W0}(i) \cdot F_{WC} \tag{8.2}$$

where

$C_{W0}(i)$: radioactivity concentration of nuclide i in the cleared items [Bq/g], and
F_{WC}: weight ratio of the cleared items in the wastes.

The radioactivity concentration $C_m(i)$ of nuclide i in the metal products made by recycling the cleared metals is derived, as shown in Eq. (4.2) in Chap. 4, from

$$C_m(i) = C_{WM}(i) \cdot F_{MC} \cdot T_i(i).G_M \cdot \exp\left(-\lambda_i t_{pd}\right) \tag{8.3}$$

where

$C_{WM}(i)$: radioactivity concentration of nuclide i in the cleared metals [Bq/g],
F_{MC}: weight ratio of the cleared metals in the metals to be recycled,
$T_i(i)$: transfer factor of element i from the recycled metals to the metal products in the melting process,
G_M: dilution factor for the recycled metals, and
t_{pd}: duration from clearance to recycling [y].

The radioactivity concentration $C_m(i)$ of nuclide i contained in the construction materials such as wall materials made by using the recycled coarse aggregate from the cleared concrete is derived as

$$Cm(i) = C_{WC}(i) \cdot F_{CC} \cdot G_C \cdot F_{RC} \cdot (F_G/\rho_C) \cdot \exp\left(-\lambda_i t_{pd}\right) \tag{8.4}$$

where

$C_{WC}(i)$: radioactivity concentration of nuclide i in the cleared concrete [Bq/g],
F_{CC}: weight ratio of the cleared concrete in the concrete to be recycled,
G_C: dilution factor for the recycled coarse aggregate,
F_{RC}: ratio of the recycled coarse aggregate to the coarse aggregate,
F_G: ratio of the coarse aggregate in the construction materials [g/m^3], and
ρ_C: density of the construction material [g/m^3].

The ratio of the recycled coarse aggregate to the coarse aggregate, F_{RC}, is the mixing ratio of the recycled coarse aggregate produced from the concrete to be recycled in the coarse aggregate as shown in Fig. 4.4. The ratio of the coarse aggregate in the construction materials, F_G, is derived by multiplying the volumetric ratio of the coarse aggregate in the construction materials by the density of the coarse aggregate.

The radioactivity concentration $C_m(i)$ of nuclide i in the parking lot base course materials made by using the recycled coarse aggregate produced by recycling the cleared concrete can be derived, as shown in Eq. (4.3), as

$$C_m(i) = C_{WC}(i) \cdot F_{CC} \cdot G_C \cdot F_{RA} \cdot \exp\left(-\lambda_i t_{pd}\right) \tag{8.5}$$

where

F_{RA}: ratio between the bitumen and the recycled coarse aggregate.

8.4.2 *Inhalation Intake Exposure Dose Evaluation Model*

Dust generated by products that have been recycled or reused, buried cleared items or by contamination attributable to cleared items can be inhaled by local residents and workers. Internal exposure doses in such cases are derived as follows.

Dust generated by cleared items is evaluated by the mass concentration method. In this method, the dust concentration that can be inhaled from the air is directly given as a scenario parameter, and the internal exposure dose $D_{INH}(i)$ [Sv/y] due to the inhalation intake of nuclide i is derived as

$$D_{INH}(i) = C_m(i) \cdot f \cdot B \cdot t_f \cdot D_{CF,INH}(i) \cdot \frac{1 - \exp(-\lambda_i t_i)}{\lambda_i t_i} \tag{8.6}$$

where

$C_m(i)$: radioactivity concentration of nuclide i in cleared items [Bq/g],
f: dust concentration in the air [g/m^3],
B: breathing rate of an adult [m^3/y],
t_f: annual exposure time during the exposure scenario, and
$D_{CF,INH}(i)$: dose conversion factor for inhalation intake of nuclide i [Sv/Bq].

Dust generated by contamination attributable to cleared items is evaluated by using the resuspension factor method. In this method, the radioactivity concentration of dust that can be inhaled from the air is derived from surface contamination density, the ratio of resuspendable loose contamination and the resuspension factor, and then the internal exposure dose $D_{INH}(i)$ due to the inhalation intake of nuclide i is derived as

$$D_{INH}(i) = C_S(i) \cdot F_l \cdot f_S \cdot B \cdot t_f \cdot D_{CF,INH}(i) \cdot \frac{1 - \exp(-\lambda_i t_i)}{\lambda_i t_i} \tag{8.7}$$

where

$C_S(i)$: surface contamination density of nuclide i of cleared items [Bq/m^2],
F_l: ratio of resuspendable loose contamination, and
f_S: resuspension factor [m^{-1}].

The radioactivity concentration of dust in the air generated by melting is derived taking into account ratio of elemental transfer to the air during the melting process and the apparent degree of concentration.

8.4.3 Ingestion Intake Exposure Dose Evaluation Model

As mentioned in Sect. 4.2.2, three types of ingestion intake are considered:

1. ingestion intake of loose contaminants from reused items;
2. ingestion intake of radioactive materials leached out of recycled metal products; and
3. ingestion intake of well water and agricultural, livestock and aquatic products containing nuclides

8.4.3.1 Ingestion Intake of Loose Contaminants

The internal exposure dose $D_{ING}(i)$ [Sv/y] in the case where a contaminant detached from the surface of a reused item attaches to the fingers of a worker and the loose contaminant is taken orally by mistake can be derived as

$$D_{ING}(i) = C_S(i) \cdot F_l \cdot F_r \cdot t_f \cdot D_{CF,ING}(i) \tag{8.8}$$

where

$C_S(i)$: surface contamination density of nuclide i on reused items [Bq/m^2],
F_l: ratio of resuspendable loose contamination,
F_r: ratio of ingestion intake of loose contamination [m^2/y],
t_f: annual exposure time during the exposure scenario, and
$D_{CF,ING}$: dose conversion factor for ingestion intake of nuclide i [Sv/Bq].

8.4.3.2 Ingestion Intake of Radioactive Material Leached Out of Recycled Metal Products

The internal exposure dose $D_{ING}(i)$ [Sv/y] in the case where radioactive material leached out of metal products manufactured from recycled metal is taken orally is derived, as shown in Eq. (4.4), as

$$D_{ING}(i) = C_m(i) \cdot A_f \cdot t_f \cdot R_e \cdot \rho_e \cdot \frac{1 - \exp(-\lambda_i t_i)}{\lambda_i t_i} \cdot D_{CF,ING}(i) \tag{8.9}$$

where

$C_m(i)$: radioactivity concentration of nuclide i in the metal product [Bq/g],
A_f: surface area of the metal product [m^2],
t_f: annual exposure time during the exposure scenario,
R_e: metal corrosion rate [m/y],
ρ_e: density of the metal [g/m^3], and
$D_{CF,ING}(i)$: dose conversion factor for ingestion intake of nuclide i [Sv/Bq].

For the annual exposure time during the exposure scenario t_f, the ratio of time of use of the metal product in a year is used.

8.4.3.3 Ingestion Intake of Well Water and Agricultural, Livestock and Aquatic Products Containing Nuclides

For evaluation pathways due to mixed wastes consisting of cleared items and non-radioactive wastes buried at the waste disposal facility, the internal exposure dose is derived by assuming that mixed wastes are disposed of at the waste disposal facility and nuclide leakage into an aquifer begins immediately after disposal. Figure 8.1 illustrates the concept of the migration of nuclides buried at the waste disposal facility.

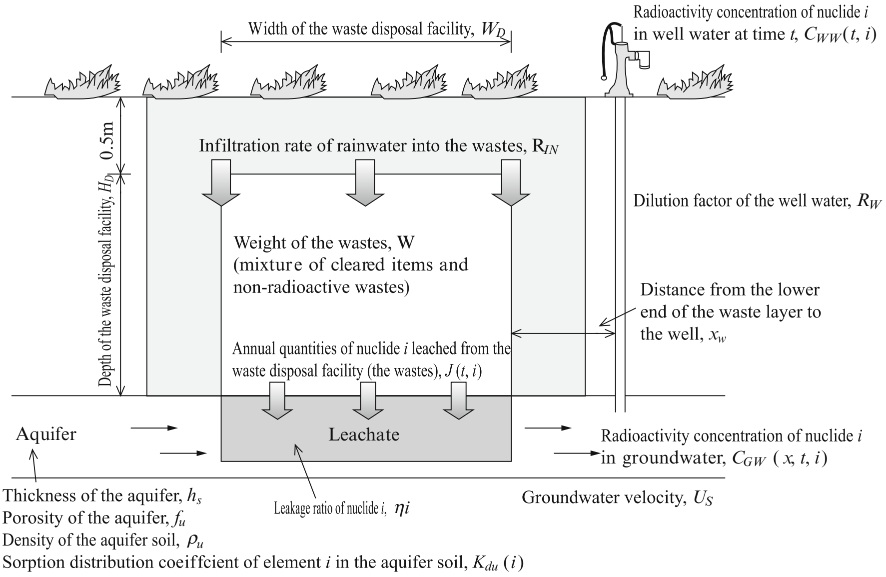

Fig. 8.1 Concept of nuclide migration due to burial disposal

As shown, nuclide i that migrates with groundwater through the aquifer flows into the well located downstream from the waste disposal facility. The internal exposure dose $D_{ING}(i)$ [Sv/y] in the case where well water is drunk is derived as

$$D_{ING}(i) = C_{WW}(t,i) \cdot Q_w \cdot D_{CF,ING}(i) \tag{8.10}$$

where

$C_{WW}(t, i)$: radioactivity concentration of nuclide i in well water at time t [Bq/m^3],
Q_w: annual drinking water intake [m^3/y], and
$D_{CF,ING}(i)$: dose conversion factor for ingestion intake of nuclide i [Sv/Bq].

The internal exposure dose $D_{ING}(i)$ [Sv/y] caused by ingesting the nuclide i contained in agricultural crops produced directly over the waste disposal facility is derived as

$$D_{ING}(i) = \sum_n C_n(i) \cdot Q_n \cdot G_n \cdot \exp(-\lambda_i t_n) \cdot D_{CF,ING}(i) \tag{8.11}$$

where

$C_n(i)$: radioactivity concentration of nuclide i in agricultural crop n [Bq/g],
Q_n: intake rate of agricultural crop n [g/y],
G_n: dilution factor for agricultural crop n,
λ_i: decay constant for nuclide i [y^{-1}],
t_n: duration of transport for agricultural crop n [y], and
$D_{CF,ING}(i)$: dose conversion factor for ingestion intake of nuclide i [Sv/Bq].

Internal exposure due to the intake of agricultural crops produced in the irrigated soil is evaluated by converting the $C_n(i)$ in Eq. (8.11) to the radioactivity concentration C_s (t, i) in the irrigated soil.

The radioactivity concentration C_{WW} (t, i) of nuclide i in well water is derived by using the emission coefficient model described in Sub-section (a) below, and the radioactivity concentration in the irrigated soil is derived from the equation shown in Sub-section (b).

(a) Emission coefficient model: The wastes buried at the waste disposal facility are exposed to infiltrating rainwater, and nuclides are leached into the aquifer. An emission coefficient model is used to quantify this phenomenon. Individual waste forms, however, are not considered. Instead, the waste disposal facility as a whole is regarded as a single large source of contamination, and the quantities of nuclide leached from the waste disposal facility (the wastes) per unit time is derived from

$$J(t,i) = \eta_i \cdot C_m(i) \cdot W \cdot \exp\{-(\lambda_i + \eta_i)t_j\} \quad t > 0 \tag{8.12}$$

where

$J(t, i)$: annual quantities of nuclide i leached from the waste disposal facility at time t [Bq/y],
η_i: leakage ratio of nuclide i [y^{-1}],
$C_m(i)$: radioactivity concentration of nuclide i in the wastes [Bq/g], and
W: weight of the wastes [g].

The leakage ratio η_i of nuclide i from the waste disposal facility (the wastes) is derived, by using the emission coefficient $R_c(i)$ for element i, as

$$\eta_i = \frac{R_{IN}}{H_D} \cdot R_c(i) \tag{8.13}$$

where

R_{IN}: infiltration rate of rainwater into the wastes [m/y],
H_D: depth of the waste disposal facility [m], and
$R_c(i)$: emission coefficient for element i.

The emission coefficient $R_c(i)$ is ratio between the amount of the element contained in the wastes and the amount of the element contained into the leachate migrating from the waste disposal facility (the wastes) to the aquifer. The emission coefficient is directly given as a scenario parameter. For its values, refer to Table 8.1.

It is assumed that nuclides leached from the waste disposal facility (the wastes) flow into the aquifer of the same width as the waste disposal facility (the wastes) and migrate along with the groundwater flow through the aquifer. It is also assumed that the density and porosity of the aquifer soil are uniform, the thickness h_s of the aquifer is constant, and the process can be expressed by an advection–dispersion equation assuming sorption distribution equilibrium (see Sect. 7.2.2) between the nuclides and the soil. That is, the radioactivity concentration $C_{GW}(x, t, i)$ [Bq/m^3] of nuclide i in groundwater at location x and time t can be expressed by the following governing equation:

$$\frac{\partial}{\partial t} R_s(i) \cdot C_{GW}(x,t,i) = D_x \frac{\partial^2 C_{GW}(x,t,i)}{\partial x^2} - U_s \frac{\partial C_{GW}(x,t,i)}{\partial x} - R_s(i) \cdot \lambda_i \cdot C_{GW}(x,t,i) \tag{8.14}$$

where

U_s: groundwater velocity [m/y],
D_x: dispersion coefficient in the x direction (groundwater flow direction) [m^2/y], and
$R_s(i)$: retardation factor of nuclide i in the aquifer.

Table 8.1 Parameter values necessary for clearance level derivation

Symbol	Unit	Parameter name	Value
A_f	m^2	Surface area of the metal product	7.07×10^{-2}: frying pan
B	m^3/y	Breathing rate of an adult	8410: occupation, 10512: loading, scrap transportation, landfill, construction of a house, agriculture, livestock farming, riverbank
D_d	m^2/y	Molecular diffusion coefficient	0.0315
D_x	m^2/y	Dispersion coefficient in the x direction (groundwater flow direction)	0
D_β	(Sv/m^2)/(Bq/y)	Dose conversion factor for skin exposure to beta rays	^{60}Co: 1.83×10^{-6}
D_γ	(Sv/m^2)/(Bq/y)	Dose conversion factor for skin exposure to gamma rays	^{60}Co: 1.48×10^{-7}
$D_{CF,\ ING}$ (i)	Sv/Bq	Dose conversion factor for ingestion intake of nuclide i	General public ^{90}Sr: 3.1×10^{-8}, ^{129}I: 1.1×10^{-7}
$D_{CF,\ INH}$ (i)	Sv/Bq	Dose conversion factor for inhalation intake of nuclide i	General public ^{241}Am: 4.2×10^{-5}
$D_{CF,\ EA}$ (i)	(Sv/y)/(Bq/g)	Dose conversion factor for external exposure to nuclide i (radioactivity concentration)	1.2×10^{-4}: refrigerator (^{60}Co)
			1.8×10^{-4}: bed (^{60}Co)
			1.9×10^{-3}: parking lot (^{60}Co)
$D_{CF,\ ES}$ (i)	(Sv/y)/(Bq/m^2)	Dose conversion factor for external exposure to nuclide i (surface contamination density)	6.4×10^{-9}: motor, valve (^{60}Co)
D_{mx}	m	Dispersion length in the x direction	0
f	g/m^3	Dust concentration in the air	5×10^{-4}: loading, landfill, construction of a house, agriculture, livestock farming, riverbank, 6×10^{-6}: occupation, 0: scrap transportation, 1×10^{-4}: scrap yard
f_d	–	Concentration factor	2: loading, landfill, construction of a house, unloading, scrap pretreatment, scrap melting, scrap reprocessing
F_G	g/m^3	Ratio of the coarse aggregate in the construction materials	1×10^{6}
F_l	–	Ratio of resuspendable loose contamination	0.01
F_r	m^2/y	Ratio of ingestion intake of loose contamination	8.76×10^{-1}
F_R	–	Soil retention factor for nuclide	1
f_s	m^{-1}	Resuspension factor	1×10^{-6}
f_u	–	Porosity of the soil	0.3: aquifer, irrigated soil

(continued)

Table 8.1 (continued)

Symbol	Unit	Parameter name	Value
F_{CC}	–	Weight ratio of the cleared concrete in the concrete to be recycled	0.1: loading, landfill, scrap pretreatment, melting, concrete crushing, slag, fabrication, 0.052: construction of house
F_{MC}	–	Weight ratio of the cleared metals in the metals to be recycled	0.7: unloading, scrap transportation, 0.1: others
F_{RA}	–	Ratio between the bitumen and the recycled coarse aggregate	0.25
F_{RC}	–	Ratio of the recycled coarse aggregate to the coarse aggregate	0.15
F_{WC}	–	Weight ratio of the cleared items in the wastes	0.1
G_C	–	Dilution factor for the recycled coarse aggregate	1
G_M	–	Dilution factor for the recycled metals	1
G_n	–	Dilution factor for agricultural crop *n*	1: agricultural crop, livestock product, aquaculture product, river product, marine product
H_D	m	Depth of the waste disposal facility	10
h_s	m	Thickness of the aquifer	3
$Kd_u\ (i)$	m^3/g	Sorption distribution coefficient of element *i* in the soil	Aquifer I: 1.0×10^{-6}, Co: 6.0×10^{-5}, Am: 2.0×10^{-3}
			Irrigated soil I: 2.7×10^{-5}, Co: 9.9×10^{-4}, Am: 1.1×10^{-1}
L_d	m	Layer thickness of dust loading on the skin	1.0×10^{-4}
L_D	m	Length of the waste disposal facility	200
P_K	g/m^2	Effective surface density of soil producing agricultural crop *K*	2.4×10^5
Q_n	g/y	Intake rate for agricultural crop *n*	7: chicken, 8: beef, 9: pork, 12: leafy vegetable, 16: egg (chicken), 22: fruit, 44: milk, 45: non-leafy vegetable, 71: rice, 250: invertebrate (river), 600: fish (river), 700: fish (aquaculture), 2,200: algae (sea), 8,100: invertebrate (sea)
Q_w	m^3/y	Annual drinking water intake	0.61 (adult)
$R_c\ (i)$	–	Emission coefficient for element *i*	1.2: H, 0.1: C, Cl, Tc, I, 3.0×10^{-2}: Ca, Mn, Fe, Co, Ni, Zn, Sr, Nb, 1.0×10^{-2}: Cs, 3.0×10^{-4}: Eu, Pu, Am
R_e	m/y	Metal corrosion rate	1.3×10^{-4}: iron
R_W	–	Dilution factor of the well water	0.33

(continued)

Table 8.1 (continued)

Symbol	Unit	Parameter name	Value
R_{IN}	m/y	Infiltration rate of rainwater into the wastes	0.4
S	–	Shielding factor for external radiation	0.2: occupation, 0.4: loading, landfill, 0.5: construction of a house, 0.9: driving, scrap transportation, 1: agriculture, livestock farming, unloading, scrap pretreatment, scrap melting, parking lot base coarse material
S_u	–	Degree of saturation of the soil moisture	0.2: upland field, pasture, 1: paddy field
t_1	y	Exposure time before start of the exposure scenario	10: construction of a house, 0: others
t_f	–	Annual exposure time during the exposure scenario	1.1×10^{-4}: refrigerator, rack, motorcycle, boat/ ship, desk, NC lathe, parking lot, unloading, driving, scrap pretreatment, scrap melting, fabrication, concrete recycling processing, loading, scrap transportation, landfill, 3.4×10^{-1}: bed, 1: occupation, 5.7×10^{-2}: construction of a house, agriculture, livestock farming, 2.3×10^{-2} slag work, reused item, 2.1×10^{-2}: frying pan
t_i	y	Exposure time during the exposure scenario	1
$T_i\,(i)$	–	Transfer factor of element *i* from the recycled metals to the metal products in the melting process	Co: 1.0
t_n	y	Duration of transport for agricultural crop *n*	0: agricultural crop, livestock product, aquaculture, river, marine product
t_{pd}	y	Duration from clearance to recycling	1
t_{WB}	y	Duration of migration from the point of where groundwater flows to the river or the sea	0
U_s	m/y	Groundwater velocity	1,220
V_{IK}	m/y	Quantity of irrigation water for agricultural crop *K*	1.2: upland field, pasture, 2.4: paddy field
V_{WB}	m^3/y	Flow rate of the river or the volume exchange rate of the sea	1×10^8: river, 8×10^9: seawater
W	g	Weight of the wastes	5×10^{11}
W_D	m	Width of the waste disposal facility	200
x_w	m	Distance from the lower end of the waste layer to the well	0

(continued)

Table 8.1 (continued)

Symbol	Unit	Parameter name	Value
x_{WB}	m	Distance from the lower end of the waste layer to the river or the sea	100: river
α	m	Effective soil depth	0.15
ρ	g/m^3	Density of dust loading on the skin	2.0×10^6: loading, landfill, construction of a house, 2.3×10^6: concrete crushing, 2.7×10^6: slag, 7.8×10^6: unloading, scrap pretreatment, scrap melting
ρ_C	g/m^3	Density of the construction material	2.3×10^6
ρ_e	g/m^3	Density of the metal	7.86×10^6: iron
ρ_u	g/m^3	Density of the soil	2.6×10^6: aquifer, irrigated soil

The dispersion coefficient D_x in the x direction, that is, the direction of groundwater flow, is derived as

$$D_x = D_{mx}|U_s| + D_d \tag{8.15}$$

where

D_{mx}: dispersion length in the x direction [m], and
D_d: molecular diffusion coefficient [m^2/y].

The retardation factor $R_s(i)$ of nuclide i in the aquifer can be derived, by using the sorption distribution coefficient $Kd_u(i)$ of element i, from the following equation:

$$R_s(i) = 1 + \frac{1 - f_u}{f_u} Kd_u(i) \cdot \rho_u \tag{8.16}$$

where

f_u: porosity of the aquifer,
ρ_u: density of the aquifer soil [g/m^3], and
$Kd_u(i)$: sorption distribution coefficient of element i in the aquifer soil [m^3/g].

The sorption distribution coefficient is a constant that is not nuclide-specific but element-specific.

The radioactivity concentration $C_{WW}(t, i)$ [Bq/m^3] of nuclide i in well water at time t is derived taking into consideration the mixing of the groundwater containing the nuclide and the surrounding groundwater that does not contain it. This dilution effect is given as

$$C_{WW}(t, i) = C_{GW}(x_w, t, i) \cdot R_W \tag{8.17}$$

where

x_w: distance from the lower end of the waste layer to well [m], and
R_W: dilution factor of the well water.

The dilution factor of the well water R_W is ratio of the quantity of contaminated groundwater to the total quantity of groundwater.

In the case where the nuclides leaking from the waste disposal facility (the wastes) migrate with groundwater and flow into the river or the sea instead of the well, the quantities of migration $A(t, i)$ [Bq/y] of nuclide i from the groundwater to the river or the sea at time t is derived, by regarding the downstream end of the waste disposal facility in the direction of groundwater flow as the origin, as

$$A(t,i) = f_u \cdot W_D \cdot h_s \cdot \left\{ U_s \cdot C_{GW}(x_{WB}, t, i) - D_x \frac{\partial}{\partial x} C_{GW}(x, t, i)|_{x=x_{WB}} \right\} \tag{8.18}$$

where

W_D: width of the waste disposal facility [m],
h_s: thickness of the aquifer [m], and
x_{WB}: distance from the lower end of the waste layer to the river or the sea [m].

In this case, the radioactivity concentration C_{WB} (t, i) [Bq/m^3] of nuclide i in the river water or seawater is derived from the equation shown below. It is assumed that the nuclide in the seawater is uniformly mixed in the evaluation region as a result of seawater exchange;

$$C_{WB}(t,i) = \frac{A(t,i)}{V_{WB}} \cdot \exp(-\lambda_i t_{WB}) \tag{8.19}$$

where

V_{WB}: flow rate of the river or the volume exchange rate of the sea [m^3/y], and
t_{WB}: duration of migration from the point of where groundwater flows to the river or the sea [y].

(b) Nuclide concentration in irrigated soil: It is assumed that agricultural land is irrigated with well water, and the nuclides contained in the well water are accumulated in the soil. The radioactivity concentration of nuclide i in the soil is derived from Eq. (8.20). The decay in concentration during the migration of the nuclide through the soil is also derived. The radioactivity concentration $C_I(t, i)$ of nuclide i in the irrigation water is derived, by assuming that it equals to the radioactivity concentration in the well water, from Eq. (8.17).

$$C_S(t,i) = F_R \int_0^t \frac{V_{IK} \cdot C_I(t,i)}{P_K} e^{-E_{IK}(t) \cdot t} dt \tag{8.20}$$

where

C_S (t, i): radioactivity concentration of nuclide i in the irrigated soil producing agricultural crop K at time t [Bq/g],
C_I (t, i): radioactivity concentration of nuclide i in irrigation water at time t [Bq/m^3],
V_{IK}: quantity of irrigation water for agricultural crop K [m/y],
P_K: effective surface density of soil producing agricultural crop K [g/m^2],
F_R: soil retention factor for nuclide, and
E_{IK} (i): effective decay factor for nuclide i in the soil producing agricultural crop K [y^{-1}].

The effective decay factor E_{IK} (i) for nuclide i in the soil where agricultural product K is produced is derived as

$$E_{IK}(i) = \lambda_i + R_{MSK}(i) \tag{8.21}$$

where

$R_{MSK}(i)$: decay factor in irrigated soil due to seepage of nuclide i [y^{-1}].

The decay factor $R_{MSK}(i)$, which indicates the time required for the applied well water to seep through the irrigated soil and for the nuclide in the well water to be accumulated in the agricultural product, is derived as

$$R_{MSK}(i) = \frac{R_{IN} + V_{IK}}{\alpha \cdot \{f_u \cdot S_u + (1 - f_u) \cdot \rho_u \cdot Kd_u(i)\}} \tag{8.22}$$

where

α: effective soil depth [m],
f_u: porosity of the irrigated soil,
S_u: degree of saturation of the soil moisture,
ρ_u: density of the irrigated soil [g/m^3], and
Kd_u (i): sorption distribution coefficient of element i in the irrigated soil [m^3/g].

8.4.4 Skin Exposure Dose Evaluation Model

The equivalent dose H_{sk} [(Sv/y)/(Bq/g)] for the skin in the case where radioactive material attaches to the body surface such as the hand surface as a result of contact with a recycled product made by recycling cleared concrete is derived from

$$H_{sk} = L_d \cdot \rho \cdot t_f \cdot f_d \cdot F_{CC} \cdot \exp(-\lambda_i t_1) \cdot \frac{1 - \exp(-\lambda_i t_i)}{\lambda_i t_i} \left(D_\beta + D_\gamma\right) \tag{8.23}$$

where

D_β: dose conversion factor for skin exposure to beta rays [(Sv/m^2)/(Bq/y)],
D_γ: dose conversion factor for skin exposure to gamma rays [(Sv/m^2)/(Bq/y)],
F_{CC}: weight ratio of the cleared concrete in the concrete to be recycled,
f_d: concentration factor,
L_d: layer thickness of dust loading on the skin [m],
ρ: density of dust loading on the skin [g/m^3],
t_1: exposure time before start of the exposure scenario [y],
t_f: annual exposure time during the exposure scenario [y], and
t_i: exposure time during the exposure scenario [y].

The equivalent dose for the skin due to contact with a recycled product made by recycling cleared metal is derived by replacing the weight ratio of the cleared concrete in the concrete to be recycled, F_{CC}, with the weight ratio of the cleared metals in metals to be recycled, F_{MC}. F_{CC} and F_{MC} are also called dilution factors. Since Safety Report Series No. 44 [4] states that radioactivity concentration increases as particle diameter decreases, the concentration factor f_d is determined by assuming that particles generated from radioactive materials become concentrated.

8.5 Scenario Parameters

Parameter values that are thought to be realistic for typical persons are determined, taking into consideration the social environment and forms of daily life in Japan. Parameter values to be used should be realistic and average values obtainable mainly from literature. If directly relevant literature is not available, either other evaluation values are used as guides or realistic and conservative values are selected. Table 8.1 shows main parameter values necessary for clearance level derivation. As mentioned earlier, the symbols shown in Table 8.1 may be different from those shown in other chapters of this book.

References

1. Nuclear Safety Commission of Japan, *Clearance Levels for Major Reactor Facilities* (1999) (in Japanese)
2. Radiation Safety Regulations Review Committee, Science and Technology Policy Bureau, Ministry of Education, Culture, Sports, Science and Technology, *Clearance Levels to be Stipulated in the Law Concerning Prevention from Radiation Hazards due to Radioisotopes, etc.* (2010) (in Japanese)
3. Nuclear Safety Commission of Japan, *Clearance Levels for Uranium-handling Facilities* (2009) (in Japanese)
4. International Atomic Energy Agency, *Derivation of Activity Concentration Values for Exclusion, Exemption and Clearance*, IAEA Safety Reports Series No. 44 (2005)

Chapter 9

Appendix B: Solving Diffusion Equations and Advection–Dispersion Equations for Radioactive Materials

Shinichi Nakayama

Chapter 7 explained that the transport of radionuclides affecting the performance assessment of radioactive waste disposal can be described with diffusion equations or advection–dispersion equations. This chapter shows some basic mass transfer problems incorporating diffusion, advection/dispersion, radioactive decay, and sorption retardation along with methods of solving them.

There are many classic books and papers dealing with methods of solving diffusion equations and advection–dispersion equations including Crank's *The Mathematics of Diffusion* [1], and solutions have been obtained under various conditions. Another good guide is Carslaw and Jaeger's *Conduction of Heat in Solids* [2] concerning heat conduction that can be expressed with the same kinds of mathematical formulae as diffusion phenomena.

This chapter deals only with problems that can be solved analytically. For example, the diffusion coefficient D is assumed to be a constant that does not depend on the concentration of material, time or groundwater flow velocity, and only one-dimensional problems are dealt with. Systems that can be solved analytically are very limited. If complex initial conditions or boundary conditions are involved, it may not be possible to obtain analytical solutions and there may be a need for numerical solution. For information on numerical solutions, refer to relevant publications.

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An Advanced Course in Nuclear Engineering, DOI 10.1007/978-4-431-55417-2_9

9.1 Diffusion in an Infinite Medium

Let us consider diffusion in a one-dimensional infinite medium having a unit cross-sectional area. Suppose there is an infinitely long water channel in which water is stationary. Now, a drop of ink having a weight of M [g] is carefully dropped into the water. The falling point is $x = 0$. We assume that at time $t = 0$, when the drop fell, the ink is instantaneously transformed into a flat plate having a unit area and a negligible thickness on the axis of $x = 0$. Over time, the ink is gradually diffused and diluted in the water. The microscopic movement of ink molecules like this is known as Brownian motion, and the macroscopic concentration C can be described by the following diffusion equation:

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2}, \quad -\infty < x < \infty, \quad 0 < t \tag{9.1}$$

where it is assumed that the diffusion coefficient D is not dependent on concentration. The conditions at time $t = 0$, that is, the initial conditions, can be written, by using Dirac's delta function $\delta(x)$, as

$$C(x, 0) = M\delta(x), \quad -\infty < x < \infty, \quad \int_{-\infty}^{\infty} \delta(x)dx = 1 \tag{9.2}$$

This equation indicates that the ink is present only in the vicinity of $x = 0$. As the boundary condition, it is assumed that both concentration and the concentration gradient are 0 at an infinite distance.

$$\left.\frac{\partial C}{\partial x}\right|_{x=\pm\infty} = 0, \quad C(\pm\infty, t) = 0, \quad 0 < t \tag{9.3}$$

Under these initial and boundary conditions, the governing equation, Eq. (9.1) is solved. The following sections introduce two methods of solution, namely, the method using Fourier transform and the variable separation method.

9.1.1 Solution Using Fourier Transform

The Fourier transform of the function $f(x)$ is $\bar{f}(w) = \int_{-\infty}^{\infty} f(x)e^{-iwx}dx$, and the Fourier inverse transform of the same function is $f(x) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \bar{f}(w)e^{iwx}dw$. The first step is to perform the Fourier transform of the governing equation, Eq. (9.1).

(Left hand side) $\int_{-\infty}^{\infty} \frac{\partial C}{\partial t} e^{-iwx} dx = \frac{d\overline{C}}{dt}$,
(Right hand side) $\int_{-\infty}^{\infty} D \frac{\partial^2 C}{\partial x^2} e^{iwx} dx = -Dw^2 \overline{C}(w,t)$

The right hand side was rewritten by using the boundary condition, (9.3). The Fourier-transformed governing equation is

$$\frac{d\overline{C}}{dt} = -Dw^2 \overline{C}, \quad 0 < t \tag{9.4}$$

The solution to this equation is given in the form of $\overline{C}(w,t) = A(w)e^{-Dw^2t}$. The initial condition Eq. (9.2) can be rewritten by Fourier transform as the following.

$$\overline{C}(w,0) = \int_{-\infty}^{\infty} C(x,0)e^{-iwx} dx = \int_{-\infty}^{\infty} M\delta(x) e^{-iwx} dx = M$$

Hence, $A(w) = \overline{C}(w,0) = M$. From these, the solution to the Fourier-transformed governing equation is

$$\overline{C}(w,t) = Me^{-Dw^2t}, \quad 0 \leqq t$$

By performing the inverse Fourier transform of the above, the following is obtained as the solution to the governing equation, Eq. (9.1):

$$\begin{aligned} C(x,t) &= \frac{1}{2\pi}\int_{-\infty}^{\infty} \overline{C}(w,t)e^{iwx} dw = \frac{M}{2\pi}\int_{-\infty}^{\infty} e^{-Dw^2t} e^{iwx} dw \\ &= \frac{M}{2\sqrt{\pi D t}} \exp\left(-\frac{x^2}{4Dt}\right) \quad -\infty < x < \infty, \quad 0 \leqq t \end{aligned} \tag{9.5}$$

where the integral $\int_{-\infty}^{\infty} e^{-\eta^2} d\eta = \sqrt{\pi}$ is used.

9.1.2 Solution by the Variable Separation Method

If we assume that the solution $C(x, t)$ to the differential equation (9.1) is in the form of the product of a function of x, $X(x)$, and a function of t, $T(t)$, then we have $C(x, t) = X(x)\,T(t)$. Substituting this in Eq. (9.1), we have $\frac{1}{DT}\frac{dT}{dt} = \frac{1}{X}\frac{d^2X}{dx^2}$ so that we can separate the variables. If the left hand side and the right hand side of this equation are equated to $\pm\alpha^2$, the next step is to solve two sets of ordinary differential equations: $\frac{1}{DT}\frac{dT}{dt} = \pm\alpha^2$ and $\frac{1}{X}\frac{d^2X}{dx^2} = \pm\alpha^2$. The solutions to these equations take the form $T(t) = ae^{D\alpha^2 t}$, $X(x) = be^{\alpha x} + ce^{-\alpha x}$ or $T(t) = ae^{-D\alpha^2 t}$, $X(x) = b\cos\alpha x + c\sin\alpha x$, where a, b and c are non-zero constants. Since the diffusion constant D is greater than zero, in the former form, both $T\,(t)$ and $X\,(|x|)$ diverge to infinity when $t \to \infty$ and $|x| \to \infty$. These can be eliminated because they cannot become solutions. In the latter form, the solution to Eq. (9.1) can be written, by using $T\,(t)$ and $X\,(x)$, as

$$C(x,t) = e^{-D\alpha^2 t}\{b(\alpha)\cos\alpha x + c(\alpha)\sin\alpha x\}, \quad -\infty < x < \infty, \quad 0 \leqq t.$$

Equation (9.1) is a linear homogeneous equation, and the following equation obtained by integrating the equation shown above with respect to α is also a solution to Eq. (9.1).

$$C(x,t) = \int_{-\infty}^{\infty} e^{-D\alpha^2 t}\{b(\alpha)\cos\alpha x + c(\alpha)\sin\alpha x\}d\alpha, \quad -\infty < x < \infty, \quad 0 \leqq t \tag{9.6}$$

Writing the initial condition as $\phi(x)$, we have Eq. (9.7).

$$C(x,0) = \phi(x) = \int_{-\infty}^{\infty} \{b(\alpha)\cos\alpha x + c(\alpha)\sin\alpha x\}d\alpha, \quad -\infty < x < \infty \tag{9.7}$$

This is an integral expression for the function $\phi(x)$. Hence, by using the Fourier integral formula, we have

$$b(\alpha) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \phi(\xi)\cos\alpha\xi d\xi, \quad c(\alpha) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \phi(\xi)\sin\alpha\xi d\xi.$$

Substituting this in Eq. (9.6), we obtain Eq. (9.8).

$$\begin{aligned} C(x,t) &= \int_{-\infty}^{\infty} e^{-D\alpha^2 t}\left\{\left(\frac{1}{2\pi}\int_{-\infty}^{\infty} \phi(\xi)\cos\alpha\xi d\xi\right)\cos\alpha x + \left(\frac{1}{2\pi}\int_{-\infty}^{\infty} \phi(\xi)\sin\alpha\xi d\xi\right)\sin\alpha x\right\}d\alpha \\ &= \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{-D\alpha^2 t}\int_{-\infty}^{\infty} \phi(\xi)\cos\alpha(x-\xi)d\alpha d\xi \\ &= \frac{1}{2\pi}\int_{-\infty}^{\infty} \phi(\xi)\int_{-\infty}^{\infty} e^{-D\alpha^2 t}\cos\alpha(x-\xi)d\alpha d\xi \\ &= \frac{1}{2\sqrt{D\pi t}}\int_{-\infty}^{\infty} \phi(\xi)\exp\left\{-\frac{(x-\xi)^2}{4Dt}\right\}d\xi, \quad -\infty < x < \infty, \quad 0 \leqq t \end{aligned} \tag{9.8}$$

Here, we applied the addition theorem of the trigonometric function, changed the integration order and used the integral, Eq. (9.9).

$$\int_{-\infty}^{\infty} e^{-D\alpha^2 t} \cos \alpha(x-\xi) d\alpha = \sqrt{\frac{\pi}{Dt}} \quad \exp\left\{-\frac{(x-\xi)^2}{4Dt}\right\} \tag{9.9}$$

This formula holds true only when the real part of the constant in the exponential part of the integrand is positive, but the formula is formally applied here.

The initial condition $\phi(x)$ $(= C(x, 0))$ is given by Eq. (9.2). Substituting it in Eq. (9.8), we obtain the following equation as the solution to the governing equation, Eq. (9.1).

$$C(x,t) = \frac{1}{2\sqrt{D\pi t}} \int_{-\infty}^{\infty} M\delta(\xi) \exp\left\{-\frac{(x-\xi)^2}{4Dt}\right\} d\xi = \frac{M}{2\sqrt{D\pi t}} \exp\left(-\frac{x^2}{4Dt}\right) \tag{9.10}$$
$$-\infty < x < \infty, \quad 0 \leqq t$$

Both Eqs. (9.5) and (9.10) are tentative solutions obtained by formally applying Eq. (9.9). The former was obtained by implicitly assuming that the solution has a Fourier transform, and the latter by assuming variable separation. In both cases, in order to verify finally that they are meaningful solutions, it is necessary to show that the governing equation Eq. (9.1), the initial condition equation Eq. (9.2), and the boundary condition equation Eq. (9.3) are satisfied. Because both of the solutions obtained satisfy these conditions, we can conclude that they are indeed solutions in the indicated ranges.

9.1.3 Average Migration Distance

A useful indicator of the magnitude of diffusion is the average migration distance $\bar{x}$. In the system under consideration here, however, diffusion occurs uniformly in the positive and negative directions. The average migration distance, therefore, is always 0, which is meaningless. In this case, the average square migration distance $\overline{x^2}$ is defined as the following.

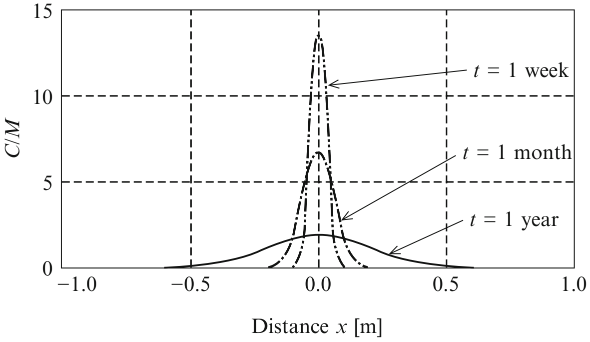
$$\overline{x^2} = \frac{1}{M} \int_{-\infty}^{\infty} x^2 C dx, \quad 0 \leqq t$$

By introducing the solution C (x, t) given by Eq. (9.5) or Eq. (9.10), we obtain Eq. (9.11).

$$\overline{x^2} = \frac{1}{2\sqrt{\pi D t}} \int_{-\infty}^{\infty} x^2 \exp\left(-\frac{x^2}{4Dt}\right) dx = \frac{4Dt}{\sqrt{\pi}} \int_0^{\infty} e^{-\eta^2} d\eta = 2Dt, \quad 0 \leqq t \tag{9.11}$$

Thus, the average migration distance is $\sqrt{\overline{x^2}} = \sqrt{2Dt}$. This means that the migration distance due to Brownian motion-induced diffusion is proportional to the square root of the time.

Fig. 9.1 Example of $\frac{C(x,\,t)}{M} = \frac{1}{2\sqrt{\pi Dt}}\exp\left(-\frac{x^2}{4Dt}\right)$



Now, let us substitute specific values and see what happens graphically to the concentration distributions expressed by Eqs. (9.5) and (9.10). As a typical value of the coefficient of diffusion D of a material in water, let us adopt 7.0×10^{-10} m^2/s. We assume that the falling point of ink is $x = 0$. The concentration distribution C/M at 1 week (7 days), 1 month (30 days) and 1 year (365 days) after falling plotted against the distance x is shown in Fig. 9.1. It can be seen that as indicated by Eqs. (9.5) and (9.10), the distributions are symmetrical with respect to $x = 0$. According to Eq. (9.11), the calculated average migration distance $\sqrt{x^2}$ at each point in time is 29 mm (1 week later), 6 cm (1 month later) and 21 cm (1 year later).

9.2 Advection in an Infinite Medium

Advection is always accompanied by dispersion (Sect. 7.2). In this section, however, we ignore dispersion, and a hypothetical system in which only advection occurs is considered. If the flow velocity of a medium in the x direction is a constant value u, the mass transfer in this medium can be expressed by the following advection equation, Eq. (9.12).

$$\frac{\partial C}{\partial t} + u\frac{\partial C}{\partial x} = 0, \quad -\infty < x < \infty, \quad 0 < t \tag{9.12}$$

Since this equation holds true if the variable is transformed as $x \to x - ut$, a general solution is given by:

$$C(x,t) = f(x - ut), \quad -\infty < x < \infty, \quad 0 < t \tag{9.13}$$

where the function f is determined by the initial condition related to $C(x, t)$. This is called an “initial value problem” or a “Cauchy problem” in honor of the mathematician who studied this problem. Because the initial condition in this particular case is Eq. (9.2), the solution to the advection equation, Eq. (9.12), is as follows.

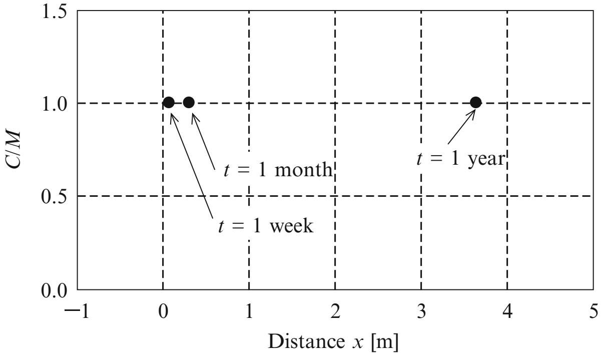


Fig. 9.2 Example of solution $\frac{C(x,t)}{M} = \delta(x - ut)$ to Eq. (9.14)

$$C(x,t) = M\delta(x - ut), \quad -\infty < x < \infty, \quad 0 \leqq t \tag{9.14}$$

If the flow velocity u of a medium is $u = 1$ cm/day, the travel distances after 1 week (7 days), 1 month (30 days) and 1 year (365 days) are 7 cm, 30 cm and 3.65 m, respectively, as shown in Fig. 9.2.

9.3 Advection and Dispersion in an Infinite Medium

This section deals with mass transfer associated with advection and dispersion. The governing advection–dispersion equation is Eq. (9.15).

$$\frac{\partial C}{\partial t} + u\frac{\partial C}{\partial x} = D\frac{\partial^2 C}{\partial x^2}, \quad -\infty < x < \infty, \quad 0 < t \tag{9.15}$$

The initial condition is Eq. (9.2), i.e., $C(x, 0) = M\delta(x)$ $(-\infty < x < \infty)$, and the boundary condition is Eq. (9.3), i.e., $\partial C/\partial t|_{x=\pm\infty} = 0$, $C(\pm\infty, 0) = 0$ $(0 < t)$.

By performing the variable transformation $\xi = x - ut$, Eq. (9.15) can be transformed as the following.

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial \xi^2}, \quad -\infty < \xi < \infty, \quad 0 < t \tag{9.16}$$

The solution to this differential equation is, as obtained from Eq. (9.8),

$$C(\xi,t) = \frac{1}{2\sqrt{D\pi t}}\int_{-\infty}^{\infty}\phi(\varsigma)\exp\left\{-\frac{(\xi-\varsigma)^2}{4Dt}\right\}d\varsigma, \quad -\infty < \xi < \infty, \quad 0 \leqq t \tag{9.17}$$

Since the initial condition is $C(x, 0) = M\delta(x) = \phi(x)$ $(-\infty < x < \infty)$, $C(\xi, t)$ can be written as

$$C(\xi,t)=\frac{1}{2\sqrt{D\pi t}}\int_{-\infty}^{\infty}M\delta(\varsigma)\exp\left\{-\frac{(\xi-\varsigma)^2}{4Dt}\right\}d\varsigma=\frac{M}{2\sqrt{D\pi t}}\exp\left(-\frac{\xi^2}{4Dt}\right),$$
$$-\infty<\xi<\infty,\quad 0\leqq t$$

Returning to the transform $\xi = x - ut$, we obtain Eq. (9.18).

$$C(x,t)=\frac{M}{2\sqrt{D\pi t}}\exp\left\{-\frac{(x-ut)^2}{4Dt}\right\},\ -\infty<x<\infty, 0\leqq t \tag{9.18}$$

By comparison with the case without the advection term, that is, the solution (9.5) or (9.10) to the diffusion equation, we can imagine a distribution in which the concentration distribution spreads out because of diffusion and there is a peak at a location deviating by $x = ut$. This is illustrated in Fig. 9.3.

Figure 9.3 shows the calculation results obtained by assuming, as in the cases shown earlier, that the diffusion coefficient D is 7.0×10^{-10} m^2/s and the ground water flow velocity u is 1 cm/day. For reference only, the lower half of Fig. 9.3 shows changes over time in concentration at different distances ($x = 0.2$ m, $x = 2.0$ m). This figure, too, can be drawn by using Eq. (9.18). The peak occurs on day 20 at $x = 0.2$ m and on day 200 at $x = 2.0$ m.

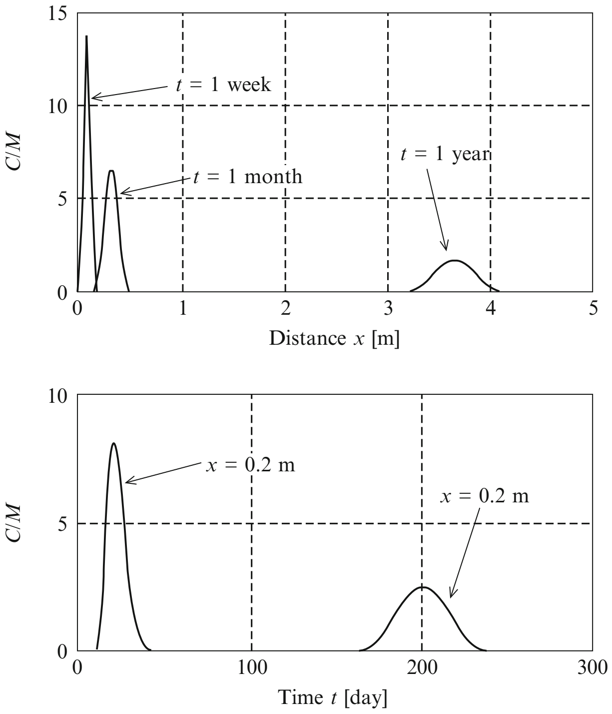


Fig. 9.3 Example of solution (9.18) to the advection–dispersion equation (*top*: concentration distribution relative to distance, *bottom*: concentration distribution relative to time)

9.4 Advection and Dispersion in an Infinite Medium Under the Retardation Effect

As explained in Sect. 7.2, if material moves while undergoing sorption to the solid phase, the retardation effect due to sorption is modeled and incorporated.

While the concentration of material in groundwater is represented by C [Bq/m^3], the concentration of material sorbed to the solid phase is represented by S [Bq/kg]. In this case, if it is assumed that the solid phase does not move, the pore distribution is uniform and porosity is constant, then the equation describing the movement of material is

$$\frac{\partial \varepsilon C}{\partial t}+\frac{\partial (1-\varepsilon)\rho S}{\partial t}+u\frac{\partial \varepsilon C}{\partial x}=D\frac{\partial^2 \varepsilon C}{\partial x^2}, \quad -\infty < x < \infty, \quad 0 < t \tag{9.19}$$

where ε is the fraction of volume occupied by pores in the solid phase, that is, porosity, and ρ [m^3/kg] is the apparent density. It may be assumed that all pores below the groundwater table are filled with groundwater.

Assume that S [Bq/kg] is proportional to the concentration C [Bq/m^3] of dissolved substances in the water phase in contact with the solid phase. The proportionality coefficient in this case is the distribution coefficient K_d [kg/m^3]. Substituting $S = K_d C$ in Eq. (9.19), we obtain

$$\frac{\partial C}{\partial t}+\frac{u}{R}\frac{\partial C}{\partial x}=\frac{D}{R}\frac{\partial^2 C}{\partial x^2}, \quad -\infty < x < \infty, \quad 0 < t \tag{9.20}$$

Where

$$R = 1+\frac{1-\varepsilon}{\varepsilon}\rho K_d \tag{9.21}$$

is called the retardation factor. If the sorption of the substances in the water phase to the solid phase does not occur, then $K_d = 0$, and therefore $R = 1$. If sorption occurs even by a small amount, then $R > 1$. The retardation factor is a parameter expressing the speed of movement of a particular substance relative to the flow velocity of groundwater. As can be seen from Eq. (9.20), the retardation factor expresses the degree of retardation of movement of a substance, specifically at u/R [m/s] compared with the groundwater flow velocity u [m/s] and at D/R [m^2/s] compared with the diffusion factor D [m^2/s].

The boundary condition for the governing equation, Eq. (9.20), is Eq. (9.3) as for the other equations described earlier. Care must be taken, however, about the initial condition because sorption to the solid phase is taken into consideration. Unlike $C(x, 0) = M\delta(x)$ for Eq. (9.2), the initial condition in this case is Eq. (9.22).

$$C(x,0) = \frac{M}{\varepsilon R}\delta(x), \qquad -\infty < x < \infty \tag{9.22}$$

This equation indicates that part of the total quantity of the substance under consideration is sorbed to the solid phase, and the remainder, $M/\varepsilon R$, is dissolved in the groundwater to become the source at $t = 0$. The coefficient εR can be understood if we consider again the definition of the retardation factor.

What is assumed in Eq. (9.22) is instantaneous equilibrium of sorption and desorption. This assumption is valid if the rates of sorption and desorption are sufficiently higher than the rates of other competing processes such as advection and dispersion. This assumption is valid at great depths at which a geological disposal facility is usually constructed or in engineered barriers because the rate of mass transfer due to advection and dispersion is sufficiently low. If, however, the sorption–desorption reaction proceeds in stages and there is a slow stage or if there are two or more chemical species in both the water and solid phases and they reach equilibrium on different time scales, the assumption of instantaneous equilibrium is not valid. In such cases, it is necessary to take kinetics into consideration.

If the assumption of instantaneous equilibrium is valid with respect to sorption and desorption, the solution to the equation of advection and dispersion, (9.20), in an infinite medium under the retardation effect can be calculated, by referring to Eq. (9.18), as follows.

$$C(x,t) = \frac{M}{2\varepsilon\sqrt{\pi D R t}} \exp\left[-\frac{\{x-(u/R)t\}^2}{4(D/R)t}\right], \qquad -\infty < x < \infty, \quad 0 \leqq t \tag{9.23}$$

The retardation effect was dealt with in an exercise in Chap. 7. In the exercise, the retardation factor R of 4.3×10^3 was obtained by using the distribution coefficient K_d of 0.4 kg/m^3 and typical values of porosity and solid phase density. This means that the rate of migration of this substance may be thought to be 1/4,300 of the groundwater flow velocity. An example calculation in this case is shown in Fig. 9.4. The example shown in Fig. 9.3 is a result obtained in a no-retardation case.

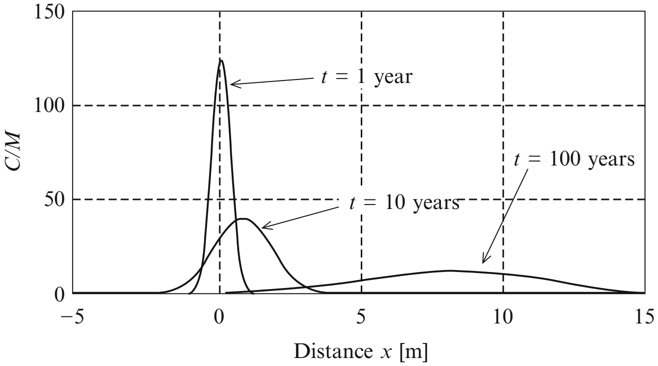


Fig. 9.4 Example calculation of a solution to a retarded material migration problem (9.23)

We note that in Fig. 9.4, the unit of time used for the curves is the year. In this case, the concentration peak, which moves by 3.65 cm in a year if there is no retardation effect, moves by only 0.1 cm under the retardation effect.

9.5 Advection and Dispersion in an Infinite Medium Accompanied by Radioactive Decay and Retardation

Next, let us consider the decay of a radioactive material. Let λ [1/s] be a decay constant. Adding the decay term λC to Eq. (9.20) gives the next governing equation.

$$\frac{\partial C}{\partial t}+\frac{u}{R}\frac{\partial C}{\partial x}=\frac{D}{R}\frac{\partial^2 C}{\partial x^2}-\lambda C, \quad -\infty<x<\infty, \quad 0<t \tag{9.24}$$

By performing the transform $C\ (x,\ t)=w(x,\ t)e^{-\lambda t}$ of the above equation, we obtain an equation of the same type as Eq. (9.20).

$$\frac{\partial w}{\partial t}+\frac{u}{R}\frac{\partial w}{\partial x}=\frac{D}{R}\frac{\partial^2 w}{\partial x^2}, \quad -\infty<x<\infty, \quad 0<t \tag{9.25}$$

As discussed in connection with Eq. (9.22), the initial condition is the following.

$$w(x,0)=\frac{M}{\varepsilon R}\delta(x), \quad -\infty<x<\infty \tag{9.26}$$

Because the solution $w(x,\ t)$ to Eq. (9.25) is in the form of Eq. (9.17) and because the solution to Eq. (9.24) is $C\ (x,\ t)=w\ (x,\ t)e^{-\lambda t}$, we obtain Eq. (9.27).

$$\begin{aligned}C(x,t)&=\frac{1}{2\sqrt{(D/R)\pi t}}\int_{-\infty}^{\infty}\frac{M}{\varepsilon R}\delta(\varsigma)\exp\left\{-\frac{(\xi-\varsigma)^2}{4(D/R)t}\right\}d\varsigma\times e^{-\lambda t}\\&=\frac{M}{2\varepsilon\sqrt{DR\pi t}}\exp\left[-\lambda t-\frac{\{x-(u/R)t\}^2}{4(D/R)t}\right],\\&-\infty<x<\infty, \quad 0\leqq t\end{aligned} \tag{9.27}$$

The difference from Eq. (9.23) is the addition of the decay term $e^{-\lambda t}$.

9.6 Diffusion in an Infinite Medium with Stepwise Initial Concentration Distribution

Suppose there is a water basin of infinite length, and the basin is divided by a partition into two sections at the point $x=0$. The water in one of the two sections (the section at $-\infty<x\leq 0$) contains a constant concentration C_0 of a substance.

The water in the other section (the section at $0 < x < +\infty$) does not contain the substance initially. The initial condition and the boundary condition for this system can be written, respectively, as the following.

$$C(x,0) = \begin{cases} 0, & 0 < x \\ C_0, & x \leqq 0 \end{cases}$$
$$C(x,t) = 0, \quad x = \infty, \quad 0 < t$$

If the partition is removed at the instant $t = 0$, the substance begins to spread out in the positive direction of x. The concentration of the substance in this case can be calculated, by using the solution with respect to a plate-shaped source dealt with in Sect. 9.1 “Diffusion in an infinite medium” as described below.

As the substance in a thin piece having a thickness of $\delta\xi$ located at $x = -\xi$ ($\xi > 0$) spreads out, the concentration at time t is, according to Eq. (9.10),

$$\frac{C_0 \delta\xi}{2\sqrt{D\pi t}} \exp\left\{-\frac{(x+\xi)^2}{4Dt}\right\}$$

and its superposition is the solution to this problem:

$$C(x,t) = \frac{C_0}{2\sqrt{D\pi t}} \int_0^\infty \exp\left\{-\frac{(x+\xi)^2}{4Dt}\right\} d\xi = \frac{1}{2} C_0 \operatorname{erfc}\left(\frac{x}{2\sqrt{Dt}}\right), \qquad (9.28)$$
$$0 < x < \infty, \quad 0 \leqq t$$

where erfc (z) ($0 \leq z$) is a complementary error function defined, along with the error function erf(z), as

$$\operatorname{erf}(z) \equiv \frac{2}{\sqrt{\pi}} \int_0^z e^{-\eta^2} d\eta, \quad \operatorname{erfc}(z) \equiv 1 - \operatorname{erf}(z)$$
$$\operatorname{erf}(-z) = -\operatorname{erf}(z), \quad \operatorname{erf}(0) = 0, \quad \operatorname{erf}(\infty) = 1$$

Figure 9.5 shows an example calculation based on the diffusion coefficient D of 7.0×10^{-10} m^2/s. In the case of a radioactive material, the right hand side of Eq. (9.28) is multiplied by a decay term, $e^{-\lambda t}$.

The equation derived here is a solution applicable to $-\infty < x < \infty$, that is, an infinite medium. A solution to the diffusion equation for a semi-infinite medium ($0 < x < \infty$) is derived in Sect. 9.9 “Diffusion in a semi-infinite medium.” A comparison may produce interesting results.

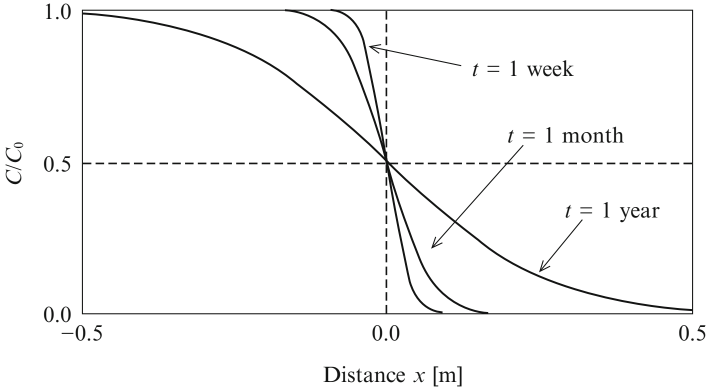


Fig. 9.5 Example calculation of $C(x,t) = \frac{1}{2}C_0 \mathrm{erfc}\left(\frac{x}{2\sqrt{Dt}}\right)$

9.7 Diffusion Under Patch-Source Condition

The problem described in Sect. 9.6 assumes that a source of infinite length exists in the negative direction of x. A little more realistic approach is to assume a source of finite extent. In Sect. 9.1 “Diffusion in an infinite medium,” a membrane-like plate source having an infinitesimally small thickness was considered. In this section, a patch source of having a finite length is considered. This means that there is initially ($t = 0$) a source distributed within the range from $x = -l$ to $x = +\, l$ at a uniform concentration C_0. The concentration $C(x, t)$ at distance x and time t is given by the following equation [1].

$$C(x,t) = \frac{1}{2}C_0\left\{\mathrm{erf}\left(\frac{l-x}{2\sqrt{Dt}}\right) + \mathrm{erf}\left(\frac{l+x}{2\sqrt{Dt}}\right)\right\}, \quad -\infty < x < \infty, \quad 0 \leqq t \quad (9.29)$$

In the case of a radioactive material, the right hand side is multiplied by a decay term, $e^{-\lambda t}$.

9.8 Advection–Dispersion Equation for a Multi-member Decay Chain

Equation (9.24) dealt with radioactive decay. Nuclides considered in connection with the equation were independent radionuclides that are not included in a decay chain. Here we look at the transition of nuclides in a decay chain. For the ith nuclide in the decay chain $A(i = 1) \rightarrow B(i = 2) \rightarrow C(i = 3) \rightarrow \ldots$ the concentration $C_i\,(x, t)$ is governed by

$$R_i\frac{\partial C_i}{\partial t}+u_i\frac{\partial C_i}{\partial x}=D_i\frac{\partial^2 C_i}{\partial x^2}-\lambda_i R_i C_i+\lambda_{i-1}R_{i-1}C_{i-1},$$
$$i=1,2,3,\cdots,\quad \lambda_0=0,\quad 0<x,\quad 0<t \tag{9.30}$$

Compared with Eq. (9.24), this equation includes the contribution of the parent nuclide $(i-1)$ in the third term on the right hand side.

The formulation of the boundary condition for the nuclides in the decay chain is somewhat complicated. Changes over time in the number of atoms of nth nuclide, $M_i\,(t)$, in the decay chain $A(i=1)\rightarrow B\;(i=2)\rightarrow C\;(i=3)\rightarrow\ldots$ can be expressed as

$$\frac{dM_i}{dt}=-\lambda_i M_i+\lambda_{i-1}M_{i-1},\quad i=1,2,3,\cdots,\quad \lambda_0=0,\quad 0<t.$$

The numbers of atoms of the first, second and third members, $M_1\,(t)$, $M_2\,(t)$ and $M_3\,(t)$, can be calculated in turn from the following equations:

$$M_1(t)=M_1^0e^{-\lambda_1 t},$$
$$M_2(t)=\frac{\lambda_1 M_1^0}{\lambda_2-\lambda_1}e^{-\lambda_1 t}+\frac{\lambda_1 M_1^0}{\lambda_1-\lambda_2}e^{-\lambda_2 t}+M_2^0e^{-\lambda_2 t},$$
$$\begin{aligned}M_3(t)=&\frac{\lambda_1\lambda_2 M_1^0}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)}e^{-\lambda_1 t}+\frac{\lambda_1\lambda_2 M_1^0}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)}e^{-\lambda_2 t}\\&+\frac{\lambda_1\lambda_2 M_1^0}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)}e^{-\lambda_3 t}+\frac{\lambda_2 M_2^0}{\lambda_3-\lambda_2}e^{-\lambda_2 t}+\frac{\lambda_2 M_2^0}{\lambda_2-\lambda_3}e^{-\lambda_3 t}+M_3^0e^{-\lambda_3 t},\\&0\leqq t\end{aligned}$$

where M_1^0, M_2^0 and M_3^0 are the quantities of the first, second and third members present initially. In general, the number of atoms for the ith nuclide is given by the formula derived by Bateman [3]:

$$M_i(t)=M_1^0\sum_{j=1}^{i}B_{ij}e^{-\lambda_j t},\quad B_{ij}\equiv\sum_{m=1}^{j}\left(\frac{M_m^0}{M_1^0}\right)\frac{\prod_{l=m}^{i}(\lambda_l/\lambda_i)}{\prod_{\substack{l=m\\ l\neq j}}^{i}\left(\lambda_l-\lambda_j\right)},\quad 0\leqq t \tag{9.31}$$

By using this formula, one of the boundary conditions for the governing equation Eq. (9.30) can be expressed as Eq. (9.32).

$$C_i(0,t)=C_1^0\sum_{j=1}^{i}B_{ij}e^{-\lambda_j t},\quad 0<t \tag{9.32}$$

The other boundary condition is that the concentration is

$$C_i(\infty,t)=0,\quad 0<t \tag{9.33}$$

at $x = \infty$, and the initial condition is that the concentration is 0:

$$C_i(x, 0) = 0, \quad 0 < x \tag{9.34}$$

This problem is solved by using Laplace transform. In Laplace transform, $f(x)$, which is a function of positive values of t, is manipulated as follows:

$$\bar{f}(p) = \int_0^\infty e^{-pt} f(t) dt \tag{9.35}$$

Some Laplace transform pairs are shown in Sect. 9.14.

The first step is to perform Laplace transform with respect to t. The first term on the left hand side of Eq. (9.30) is

$$\int_0^\infty e^{-pt} R_i \frac{\partial C_i}{\partial t} dt = \left[R_i C_i(x,t) e^{-pt}\right]_0^\infty + p \int_0^\infty R_i C_i(x,t) e^{-pt} dt = pR_i \overline{C_i}(x,p) \tag{9.36}$$

Here, we use Eq. (9.33) and the condition that the exponential term becomes 0 when $t = \infty$. The Laplace-transformed governing equation is the next expression.

$$D_i \frac{d^2 \overline{C_i}}{dx^2} - u_i \frac{d \overline{C_i}}{dx} - (p + \lambda_i) R_i \overline{C_i} + \lambda_{i-1} R_{i-1} \overline{C_{i-1}} = 0 \tag{9.37}$$

The Laplace-transformed boundary condition, Eq. (9.32), is given as the following.

$$\overline{C_i}(0,p) = \int_0^\infty e^{-pt} C_1^0 \sum_{j=1}^{i} B_{ij} e^{-\lambda_j t} dt = C_1^0 \sum_{j=1}^{i} B_{ij} \frac{1}{p + \lambda_i} \tag{9.38}$$

Since no general solution to the advection–dispersion equation, Eq. (9.30), can be found for the ith nuclide in the decay chain, a solution is found one by one starting from the parent nuclide $i = 1$. The solution $C_i(x, t)$ ($i = 1, 2, 3$) for $i = 1, 2, 3$ is shown in Ref. [4]. The same reference also show solutions for not only a semi-infinite medium ($0 < x$), as for the governing equation (9.30), but also an infinite medium ($-\infty < x < \infty$).

9.9 Diffusion in a Semi-infinite Medium

The preceding sections have looked at a pipe-like medium of infinite length. In such a medium, substances spread in both the positive and negative directions. In this section, a semi-infinite medium in which diffusion occurs only in the positive direction of x is considered. The diffusion equation, whose domain differs from that of Eq. (9.1) for an infinite medium, is written as

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2}, \quad 0 < x, \quad 0 < t \tag{9.39}$$

As the boundary condition, it is assumed that concentration is always kept at C_0 at the point $x = 0$:

$$C(0, t) = C_0, \quad 0 < t \tag{9.40}$$

As the initial condition, it is assumed that the substance under consideration was not present in the region defined as $0 < x$:

$$C(x, 0) = 0, \quad 0 < x \tag{9.41}$$

By performing the Laplace transform of the governing equation, Eq. (9.39), we can rewrite the left hand side as

$$\int_0^\infty e^{-pt}\frac{\partial C}{\partial t}dt = [C(x,t)e^{-pt}]_0^\infty - (-p)\int_0^\infty C(x,t)e^{-pt}dt = p\overline{C}(x,p) - C(x,0).$$

Here, from the initial condition Eq. (9.41), we used the conditions that $C(x, 0) = 0$ and that the exponential term becomes 0 if $t = \infty$. By performing the Laplace transform of the right hand side of Eq. (9.39), we obtain the following.

$$\int_0^\infty e^{-pt}\frac{\partial^2 C}{\partial x^2}dt = \frac{d^2}{dx^2}\int_0^\infty e^{-pt}C(x,t)dt = \frac{d^2}{dx^2}\overline{C}(x,p)$$

Hence, the governing equation, Eq. (9.39), can be Laplace-transformed as Eq. (9.42).

$$D\frac{d^2\overline{C}}{dx^2} = p\overline{C} \tag{9.42}$$

The solution to this equation can be expressed as a linear combination of the term $\exp\left(\sqrt{p/Dx}\right)$ and $\exp\left(-\sqrt{p/Dx}\right)$. Divergence, however, is not physically permissible at $x = \infty$, and it is necessary to satisfy the equation derived by performing the Laplace transform of the boundary equation, (9.40):

$$\overline{C}(0,p) = \int_0^\infty C_0 e^{-pt}dt = \frac{C_0}{p} \tag{9.43}$$

The solution, therefore, to the Laplace-transformed diffusion equation, Eq. (9.42) is

$$\overline{C}(x,p) = \frac{C_0}{p}\exp\left(-\sqrt{\frac{p}{D}}x\right) \tag{9.44}$$

The equation shown below, which is derived by performing the inverse Laplace transform of the above equation, is the solution to Eq. (9.39). Here, we applied the condition that the inverse Laplace transform of $e^{-a\sqrt{p}}/p(a > 0)$ is $\mathrm{erfc}\left(a/2\sqrt{t}\right)$ (see Sect. 9.14).

$$C(x,t) = C_0\mathrm{erfc}\left(\frac{x}{2\sqrt{Dt}}\right), \quad 0 \leqq x, \quad 0 \leqq t \tag{9.45}$$

This solution is exactly two times as large as the solution (9.28) obtained for an infinite medium in Sect. 9.6 "Diffusion in an infinite medium with stepwise concentration distribution." The reader may want to study further to find out why the factor becomes 2.

9.10 Solution for a Band-Release Condition

The boundary condition equation, Eq. (9.40), shown in the preceding section indicates that a constant concentration C_0 continues to be supplied at the point $x = 0$ at and after $t = 0$. In this section, let us consider the case where supply continues for a fixed period of time T. This band-release boundary condition C^B (0, t) can be written, by using the Heaviside function H (t), as follows.

$$C^B(0,t) = C_0\{H(t) - H(t-T)\} = \begin{cases} 0, & t < 0 \\ C_0, & 0 \leqq t \leqq T \\ 0, & T < t \end{cases} \tag{9.46}$$

Hence, a boundary condition for the advection and dispersion of the ith nuclide in a decay chain like the one dealt with in Sect. 9.8 "Advection–dispersion equation for a multi-member decay chain" is expressed as

$$C_i^B(0,t) = C_1^0\sum_{j=1}^{i} B_{ij}e^{-\lambda_j t}\{H(t) - H(t-T)\}, \quad B_{ij} = \sum_{m=1}^{j}\left(\frac{C_m^0}{C_1^0}\right)\frac{\prod_{l=m}^{i}(\lambda_l/\lambda_i)}{\prod_{\substack{l=m \\ l \neq j}}^{i}(\lambda_l/\lambda_j)} \tag{9.47}$$

It has been found that solutions under band-release boundary conditions can be obtained by the superposition of solutions under the condition $0 \leq t$ and solutions under the condition $T \leq t$ [5]. The solution C_i^B (x, t) to the governing equation, Eq. (9.30), therefore, can be calculated as follows:

$$C_i^B(x,t) = C_i\left(x, t : B_{ij}\right) - C_i\left(x, t - T : B_{ij}e^{-\lambda_i T}\right).$$

9.11 Diffusion in a Finite Medium

The preceding sections have considered the movement of material in an infinite or semi-infinite medium. Finally, this section deals with diffusion in a finite medium. Since an infinite medium and a finite medium have different boundary conditions, solutions obtained differ from the ones obtained thus far.

The diffusion coefficient for ions in pore water in rocks or in the bentonite buffer material used at disposal facilities can be determined under laboratory conditions by using the method called the through-diffusion method. The analysis of measurement results obtained by the through-diffusion method requires a diffusion equation expressing material movement in a finite medium. In Exercise 2 of Chap. 7, the diffusion coefficient for cesium (Cs) was determined by using the solution to the diffusion equation. Let us use that solution here.

A through-diffusion experiment is conducted by using disc-shaped bentonite-sand mixture specimens and the experimental cell apparatus shown in Fig. 9.6. A disc-shaped specimen is placed between two cells filled with an aqueous solution. The aqueous solution in the cell on the left hand side in Fig. 9.6 contains a high concentration of Cs, while the right-hand-side cell is filled with water that does not contain Cs at first. The Cs ions in the high-concentration cell move through the disc-shaped specimen by diffusion.

It is assumed that the movement of Cs in the disc-shaped specimen follows the diffusion equation.

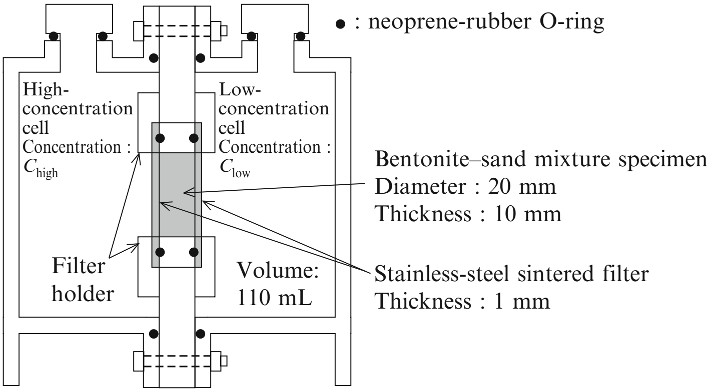


Fig. 9.6 Through-diffusion cell apparatus

$$R\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2}, \quad 0 < x < L, \quad 0 < t \tag{9.48}$$

The symbols used in connection with this problem are as follows:

C: Cs concentration in pore water in bentonite-sand mixture specimen [Bq/m^3]
D: diffusion coefficient for Cs in pore water in the specimen [m^2/s]
R: retardation factor $= 1 + (1 - \varepsilon)\rho K_d/\varepsilon$
ε: porosity of specimen
ρ: density of specimen [kg/m^3]
K_d: sorption distribution coefficient [m^3/kg]
L: thickness of disc-shaped specimen [m]
t: time [s]
x: distance in disc-shaped specimen [m], where the interface with the aqueous solution in the high-concentration cell is defined as $x = 0$, and the x-axis is defined in the direction of the low-concentration cell.

It is necessary here to be careful about the diffusion coefficient D. Diffusion in this case is not diffusion in free water. Instead, it is diffusion in rock, a porous medium. When a material spreads in pore water, it is affected by such factors as the shape of pores. Consequently, the degree of diffusion is not the same as that in free water. The coefficient of diffusion in pores tends to be smaller than the coefficient of diffusion in free water and is given as a value proportional to the diffusion coefficient D. The value obtained by multiplying the coefficient of diffusion in pore water by the porosity ε is called the effective diffusion coefficient. If the material in pore water spreads in the pore water without being sorbed to the medium, the actual diffusion coefficient is the same as the effective diffusion coefficient. If retardation due to sorption is taken into consideration, the apparent diffusion coefficient can be given as D/R as can be seen from Eq. (9.48).

Since Cs is not present in the bentonite-sand mixture specimen at $t = 0$, the initial condition for the differential equation, Eq. (9.48), is

$$C(x, 0) = 0, \quad 0 < x < L \tag{9.49}$$

The boundary condition is given as follows: Cs is put into the high-concentration cell in a quantity by far greater than the quantity expected to spread from the high-concentration cell into the specimen, and it is assumed that the Cs concentration in the high-concentration cell, $C_{\text{high}}(t)$, is kept more or less constant at the initial concentration C_0 during the experiment period. It is therefore assumed that the concentration of Cs in the pore water in the specimen at the interface ($x = 0$) on the high-concentration cell is equal to the initial concentration C_0:

$$C(0, t) = C_0, \quad 0 < t \tag{9.50}$$

The concentration on the low-concentration side, $C_{\mathrm{low}}\ (t)$, gradually rises over time from the initial value of 0, but it may be regarded approximately as 0 because it is negligibly small compared with the concentration C_0 in the high-concentration cell. The concentration, therefore, at the interface ($x = L$) between the disc-shaped specimen and the low-concentration cell is assumed to be 0:

$$C(L,t) = 0, \quad 0 < t \tag{9.51}$$

As mentioned in Sect. 9.9 "Diffusion in a semi-infinite medium," the solution to the differential equation, (9.48), under the initial condition equation, (9.49), can be expressed as a linear combination of the terms $\exp\left(\sqrt{p/Dx}\right)$ and $\exp\left(-\sqrt{p/Dx}\right)$. By using the boundary conditions, (9.50), and Eq. (9.51), the Laplace-transformed solution can be expressed as Eq. (9.52).

$$\overline{C}(x,p) = \frac{C_0}{p} \cdot \frac{\sinh q(L-x)}{\sinh qL}, \quad q = \sqrt{\frac{p}{D/R}} \tag{9.52}$$

In order to perform the inverse Laplace transform of the above equation, the following property is used. If an inversely transformed function $y(p)$ can be expressed by using two polynomials-$f\,(p)$ and $g\,(p)$- that do not have any common term in the form

$$\overline{y}(p) = \frac{f(p)}{g(p)}$$

and if the degree of the numerator $f(p)$ is smaller than the degree of the denominator $g(p)$ and the solutions a_1, a_2, . . ., a_n to the equation

$$g(p) = (p - a_1)(p - a_2)\cdots(p - a_n) = 0$$

are all different, then the original function $y(t)$ can be given by Eq. (9.53).

$$y(t) = \sum_{r=1}^{n} \frac{f(a_r)}{g'(a_r)} e^{a_r t} \tag{9.53}$$

For problems involving diffusion in a finite medium, the sum n in this equation may be deemed to be ∞ (infinity). The function sinh z can be extended as

$$\sinh z = z\left(1 + \frac{z^2}{\pi^2}\right)\left(1 + \frac{z^2}{2^2\pi^2}\right)\left(1 + \frac{z^2}{3^2\pi^2}\right)\cdots\left(1 + \frac{z^2}{n^2\pi^2}\right)\cdots.$$

In Eq. (9.52), therefore, if the common factor z is eliminated, the following expressions can be used:

$$\begin{aligned} f(p) &= C_0\frac{1}{q}\sinh q(L-x) = C_0(L-x)\prod_{m=1}^{\infty}\left[1+\frac{(q(L-x))^2}{m^2\pi^2}\right] \\ g(p) &= p\frac{1}{q}\sinh qL = pL\prod_{m=1}^{\infty}\left[1+\frac{(qL)^2}{m^2\pi^2}\right]. \end{aligned}$$

The values that give 0 to $g\ (p)$ are

$$p=-\frac{Dn^2\pi^2}{L^2R}\quad (n=1,2,\cdots),\quad p=0.$$

Substituting $p=-Dn^2\pi^2/L^2R$ (or $qL=\pm in\pi$) ($n=1, 2, \ldots$) and $p=0$ ($q=0$) in the denominator

$$g'(p)=\frac{1}{2q}(\sinh qL+qL\cosh qL),\quad q=\sqrt{\frac{p}{D/R}}$$

of Eq. (9.53), we obtain

$$\begin{aligned} g'(p)\big|_{p=-Dn^2\pi^2/RL^2,\ qL=\pm in\pi(n=1,2,\cdots)} &= \frac{\sinh(\pm in\pi)+(\pm in\pi)\cosh(\pm in\pi)}{2q} \\ &= \frac{i\sin(\pm n\pi)+(\pm in\pi)\cos(\pm n\pi)}{2q} \\ &= \frac{(\pm in\pi)(-1)^n}{2q}=\frac{(-1)^nL}{2}\quad (n=1,2,3,\cdots) \end{aligned}$$

$$g'(p)\big|_{p=0,\ q=0}=\frac{1}{2}\left(L\prod_{i=1}^{\infty}\left[1+\frac{(qL)^2}{i^2\pi^2}\right]\right)_{q=0}+\frac{1}{2}L\cosh qL=L.$$

The numerator $f\,(p)$ can be calculated as

$$\begin{aligned} f(p)\big|_{p=-Dn^2\pi^2/RL^2,\ qL=\pm in\pi(n=1,2,\cdots)} &= C_0\frac{L}{\pm in\pi}\sinh\left(\frac{\pm in\pi(L-x)}{L}\right) \\ &= C_0\frac{L}{\pm in\pi}\sin\left(\frac{\pm n\pi(L-x)}{L}\right) \\ &= C_0\frac{L}{n\pi}\sin\left(\frac{n\pi(L-x)}{L}\right) \\ &= C_0\frac{L}{n\pi}\left(\sin(n\pi)\cos\frac{n\pi x}{L}-\cos(n\pi)\sin\frac{n\pi x}{L}\right) \\ &= C_0\frac{(-1)^{n+1}L}{n\pi}\sin\frac{n\pi x}{L}\quad (n=1,2,3,\cdots) \end{aligned}$$

$$f(p)\big|_{p=0,\ q=0}=C_0(L-x).$$

By using these equations, we obtain Eq. (9.54) as the concentration distribution of Cs in the bentonite-sand mixture disc-shaped specimen that we are trying to calculate here.

$$C(x,t) = \sum_{n=1}^{\infty} \frac{f(p_n)}{g'(p_n)} e^{p_n t} = C_0\left(1 - \frac{x}{L}\right) - \frac{2C_0}{\pi}\sum_{n=1}^{\infty}\frac{1}{n}\sin\left(\frac{n\pi x}{L}\right)\exp\left(-\frac{Dn^2\pi^2}{L^2R}t\right) \quad 0\leqq x\leqq L, \quad 0\leqq t \tag{9.54}$$

The flux $J(L, t)$ of Cs diffusing into the low-concentration cell can be calculated, by using Eq. (9.54), as Eq. (9.55).

$$\begin{aligned} J(L,t) &= -D\left.\frac{\partial C(x,t)}{\partial x}\right|_{x=L} \\ &= C_0\frac{D}{L}\left[1 + 2\sum_{n=1}^{\infty}(-1)^n \exp\left(-\frac{Dn^2\pi^2 t}{L^2R}\right)\right], \quad 0\leqq t \end{aligned} \tag{9.55}$$

By using this flux, changes in the concentration in the low-concentration cell, $C_{\mathrm{low}}(t)$, can be calculated as follows:

$$\begin{aligned} \frac{C_{\mathrm{low}}(t)}{C_0} &= \frac{A}{VC_0}\int_0^t J(L,t)dt \\ &= \frac{AD}{VL}\left[t - \frac{L^2R}{6D} - \frac{2L^2R}{D\pi^2}\sum_{n=1}^{\infty}\frac{(-1)^n}{n^2}\exp\left(-\frac{Dn^2\pi^2 t}{L^2R}\right)\right], \quad 0\leqq t \end{aligned} \tag{9.56}$$

where V is the volume of the solution in the cell, and A is the area of the disc-shaped specimen surface in contact with the solution. Also, the formula

$$\sum_{n=1}^{\infty}\frac{(-1)^{n-1}}{n^2} = \frac{\pi^2}{12}$$

is used. As can be seen from Eq. (9.56), after a sufficient amount of time, the concentration begins to rise in proportion to time as follows.

$$\frac{C_{\mathrm{low}}(t)}{C_0} = \frac{AD}{VL}\left(t - \frac{L^2R}{6D}\right), \quad 0\leqq t \tag{9.57}$$

In this state, the distribution of concentration in the disc-shaped specimen does not depend on time, which means a steady state

$$\frac{C(x,t)}{C_0} = 1 - \frac{x}{L}, \quad 0\leqq x\leqq L, \quad 0\leqq t \tag{9.58}$$

has been reached.

As Eq. (9.57) indicates, the slope of this linear part is AD/LV, and the point of intersection of the extrapolated line of the linear part and the time axis, or the t-intercept, is $t = L^2R/6D$. Since A, L and V are known, the values of the diffusion coefficient D and the retardation factor R can be obtained if the values of the slope and the intercept can be determined from experimental results. The value of the distribution coefficient K_d can also be determined from R.

For Eq. (9.52), the Laplace-transformed solution has been shown by using a hyperbolic function. The solution, however, can also be written as follows.

$$\overline{C}(x,p) = \frac{C_0}{p} \cdot \frac{e^{q(L-x)} - e^{-q(L-x)}}{e^{qL} - e^{-qL}}, \quad q = \sqrt{\frac{p}{D/R}}$$

From this, a solution of a form different from that of Eq. (9.54) can be derived. By rewriting the above equation, we obtain the following.

$$\begin{aligned}\overline{C}(x,p) &= \frac{C_0}{p} \cdot \frac{e^{qx} - e^{-q(2L-x)}}{1 - e^{-2qL}} = \frac{C_0}{p}\left\{e^{-qx} - e^{-q(2L-x)}\right\}\sum_{n=1}^{\infty} e^{-2qnL} \\ &= \frac{C_0}{p}\sum_{n=1}^{\infty} e^{-q(x+2nL)} - \frac{C_0}{p}\sum_{n=1}^{\infty} e^{-q[x-(2n+1)L]}\end{aligned}$$

Since the inverse Laplace transform of $e^{-a\sqrt{p}}/p\,(a > 0)$ is $\operatorname{erfc}\left(a/2\sqrt{t}\right)$, we obtain

$$C(x,t) = C_0\sum_{n=1}^{\infty}\operatorname{erfc}\left(\frac{x+2nL}{2\sqrt{(D/R)t}}\right) - C_0\sum_{n=1}^{\infty}\operatorname{erfc}\left(\frac{x-(2n+1)L}{2\sqrt{(D/R)t}}\right), \quad 0 \leqq x \leqq L, \quad 0 \leqq t$$

which is another form of Eq. (9.54), but the equation in the present form cannot be used to analyze data obtained from through-diffusion experiments.

The analysis described thus far is supposed to be used in cases where the concentration in the high-concentration cell is kept higher than the concentration in the low-concentration cell and the concentration at the interface between the disc-shaped specimen and the aqueous solution can be approximated as in Eqs. (9.50) and (9.51) as assumed at the outset. If the substance under consideration is hardly soluble or if diffusion is so fast that such an approximation is not appropriate, it is necessary to consider exact solutions as shown below.

The governing equations for the concentration in the specimen, $C\,(x,\, t)$, the concentration in the high-concentration cell, $C_{\text{high}}\,(x,\, t)$, and the concentration in the low-concentration cell, $C_{\text{low}}(x,t)$, and the initial condition and the boundary condition, are given below.

$$\frac{\partial C}{\partial t} = \frac{D}{R}\frac{\partial^2 C}{\partial x^2}, \quad 0 < t, \quad 0 < x < L \quad C(x,0) = 0, \quad 0 < x < L$$

$$\frac{dC_{\text{high}}}{dt} = \frac{D}{R_V}\left.\frac{dC}{dt}\right|_{x=0}, \quad 0 < t \quad C_{\text{high}}(t) = C(0,t), \quad 0 < t \quad C_{\text{high}}(0) = C_0$$

$$\frac{dC_{\text{low}}}{dt} = -\frac{D}{R_V}\left.\frac{dC}{dt}\right|_{x=L}, \quad 0 < t \quad C_{\text{low}}(t) = C(L,t), \quad 0 < t \quad C_{\text{low}}(0) = 0$$

By applying Laplace transform to this differential equation, we can obtain the following solutions:

$$\begin{cases} \dfrac{C(x,t)}{C_0} = \dfrac{1}{2}\cdot\dfrac{1}{1+l} + \displaystyle\sum_{n=1}^{\infty}\dfrac{\exp\left(-\alpha_n^2 Dt/b^2\right)}{1+l+l\alpha_n^2}\cdot\dfrac{\cos\alpha_n(x/b-l)}{\cos\alpha_n l} \\ \qquad\qquad - \displaystyle\sum_{n=1}^{\infty}\dfrac{\exp\left(-\beta_n^2 Dt/b^2\right)}{1+l+l\beta_n^2}\cdot\dfrac{\sin\beta_n(x/b-l)}{\sin\beta_n l}, \\ \dfrac{C_{\text{high}}(t)}{C_0} = \dfrac{1}{2}\cdot\dfrac{1}{1+l} + \displaystyle\sum_{n=1}^{\infty}\dfrac{\exp\left(-\alpha_n^2 Dt/b^2\right)}{1+l+l\alpha_n^2} + \displaystyle\sum_{n=1}^{\infty}\dfrac{\exp\left(-\beta_n^2 Dt/b^2\right)}{1+l+l\beta_n^2}, \\ \dfrac{C_{\text{low}}(t)}{C_0} = \dfrac{1}{2}\cdot\dfrac{1}{1+l} + \displaystyle\sum_{n=1}^{\infty}\dfrac{\exp\left(-\alpha_n^2 Dt/b^2\right)}{1+l+l\alpha_n^2} - \displaystyle\sum_{n=1}^{\infty}\dfrac{\exp\left(-\beta_n^2 Dt/b^2\right)}{1+l+l\beta_n^2}, \end{cases}$$

where α_n and β_n are non-negative solutions to $\alpha_n + \tan\alpha_n l = 0$ and $\beta_n \tan\beta_n l = 1$, respectively; $l \equiv bL/2$, $b \equiv R_V/R$, $R_V \equiv V/\varepsilon A$; R is a retardation factor calculated as $R = 1 + (1 - \varepsilon)\rho K_d/\varepsilon$; V is cell capacity; and A is the area of contact between the disc-shaped specimen and the solution.

9.12 Diffusion Equation in Cylindrical Coordinate System

We have thus far dealt with one-dimensional diffusion in a Cartesian coordinate system. The diffusion equation in a three-dimensional coordinate system is as follows.

$$\frac{\partial C}{\partial t} = D\left(\frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2} + \frac{\partial^2 C}{\partial z^2}\right)$$

To transform this to a formula for a cylindrical coordinate system (r, θ, z), variables x and y are transformed to $r\cos\theta$ and $r\sin\theta$, respectively.

$$\frac{\partial}{\partial r}=\frac{\partial}{\partial x}\frac{\partial x}{\partial r}+\frac{\partial}{\partial y}\frac{\partial y}{\partial r}=\cos\theta\frac{\partial}{\partial x}+\sin\theta\frac{\partial}{\partial y},$$

$$\frac{\partial}{\partial\theta}=\frac{\partial}{\partial x}\frac{\partial x}{\partial\theta}+\frac{\partial}{\partial y}\frac{\partial y}{\partial\theta}=-r\sin\theta\frac{\partial}{\partial x}+r\cos\theta\frac{\partial}{\partial y}$$

Hence, we obtain

$$\frac{\partial}{\partial x}=\cos\theta\frac{\partial}{\partial r}-\frac{\sin\theta}{r}\frac{\partial}{\partial\theta},\quad \frac{\partial}{\partial y}=\sin\theta\frac{\partial}{\partial r}+\frac{\cos\theta}{r}\frac{\partial}{\partial\theta}$$

and

$$\frac{\partial^2}{\partial x^2}=\frac{\partial}{\partial x}\left(\cos\theta\frac{\partial}{\partial r}-\frac{\sin\theta}{r}\frac{\partial}{\partial\theta}\right),\quad \frac{\partial^2}{\partial y^2}=\frac{\partial}{\partial y}\left(\sin\theta\frac{\partial}{\partial r}+\frac{\cos\theta}{r}\frac{\partial}{\partial\theta}\right).$$

From these, we obtain

$$\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}=\frac{\partial^2}{\partial r^2}+\frac{1}{r}\frac{\partial}{\partial r}+\frac{1}{r^2}\frac{\partial^2}{\partial\theta^2}.$$

Hence, we obtain Eq. (9.59) as the diffusion equation for the cylindrical coordinate system.

$$\frac{\partial C}{\partial t}=D\left(\frac{1}{r}\frac{\partial}{\partial r}r\frac{\partial C}{\partial r}+\frac{1}{r^2}\frac{\partial^2 C}{\partial\theta^2}+\frac{\partial^2 C}{\partial z^2}\right)\tag{9.59}$$

9.13 Diffusion in Spherical Coordinate System

Let us move a step further to derive a formula for a spherical coordinate system (r, θ, ϕ). The transformation of a Cartesian coordinate system to a spherical coordinate system can be expressed as $x=r\sin\theta\cos\phi$, $y=r\sin\theta\sin\phi$ and $z=r\cos\theta$. It can also be performed through the transformation to a cylindrical coordinate system described in Sect. 9.12.

If $R=r\sin\theta$ and $z=r\cos\theta$, then the transformation becomes the same operation as the transformation from a Cartesian coordinate system to a cylindrical coordinate system expressed as $x=R\cos\phi$, $y=R\sin\phi$ and $z=z$. Then, we obtain an equation of the same form as Eq. (9.59).

$$\frac{\partial C}{\partial t}=D\left(\frac{1}{R}\frac{\partial}{\partial R}R\frac{\partial C}{\partial R}+\frac{1}{R^2}\frac{\partial^2 C}{\partial\phi^2}+\frac{\partial^2 C}{\partial z^2}\right)$$

As a next step, by calculating differential coefficients regarding R and z as functions of r and θ, we obtain the following diffusion equation for a spherical coordinate system.

$$\frac{\partial C}{\partial t} = D\left(\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial C}{\partial r}\right) + \frac{1}{r^2\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial C}{\partial\theta}\right) + \frac{1}{r^2\sin\theta}\frac{\partial^2 C}{\partial\phi^2}\right) \tag{9.60}$$

9.14 Laplace Transform Pairs

$$f(t) = \frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty} e^{pt}g(p)dp, \quad g(p) = \int_0^\infty e^{-pt}f(t)dt, \quad 0 \leqq t, \quad 0 < c$$

Table 9.1 shows Laplace transform pairs of some representative functions.

Table 9.2 shows Laplace transforms of a limited number of fundamental functions.

Table 9.1 Laplace transforms of some representative functions

Original function $f(t)$	Laplace – transformed function $g(t)$
$f(at)\,(a>0)$	$\frac{1}{a}g\left(\frac{p}{a}\right)$
$t^n f(t)$	$(-1)^n\frac{d^n g(p)}{dp^n}$
$\frac{1}{t^n}f(t)$	$\underbrace{\int_p^\infty \cdots \int_p^\infty}_{n} g(p)(dp)^n$ (n-multiple integral)
$e^{at}f(t)$	$g(p-a)$
$f'(t)$	$pg(p)-f(0)$
$f^{(n)}(t)$	$p^n g(p) - \sum_{r=0}^{n-1} p^{n-r-1}f^{(r)}(0)$
$\int_0^t f(s)ds$	$\frac{1}{p}g(p)$
$\int_0^t \cdots \int_0^t f(s)(ds)^n$ (n-multiple integral)	$\frac{1}{p^n}g(p)$
$\int_t^\infty \frac{f(s)}{s}ds$	$\frac{1}{p^n}\int_0^p g(p)dp$
$\left.\begin{matrix} f(t-a)\,(t\geq a) \\ 0\,(0\leq t<a)\end{matrix}\right\}$	$e^{-ap}g(p)$
$f(t^2)$	$\frac{1}{\sqrt{\pi}}e^{-p^2s^2/4}g\left(\frac{1}{s^2}\right)\frac{ds}{s^2}$
$\int_0^t f_1(s)f_2(t-s)ds = (f_1 * f_2)(t)$	$g_1(p)g_2(p)$
$\frac{1}{\sqrt{t}}\int_t^\infty e^{-s^2/(4t)}f(s)ds$	$\sqrt{\frac{\pi}{p}}g(\sqrt{p})$

$(f_1 * f_2)(t)$ is a convolution of $f_1(t)$ and $f_2(t)$. The Laplace transforms of $f_1(t)$ and $f_2(t)$ are $g_1(t)$ and $g_2(t)$, respectively

Table 9.2 Laplace transforms of some fundamental functions

Original function	Laplace – transformed function
$H(t) = \begin{cases} 1(t > 0) \\ 0(t < 0) \end{cases}$	$\frac{1}{p}$
$H(t-a) = \begin{cases} 1(t > a) \\ 0(t < a) \end{cases} (a > 0)$	$\frac{e^{-ap}}{p}$
e^{at}	$\frac{1}{p - \alpha}$
$\operatorname{erfc}\left(\frac{a}{2\sqrt{t}}\right) (a > 0)$	$\frac{e^{-a\sqrt{p}}}{p}$
$e^{-t^2/(4a)} (a > 0)$	$2\sqrt{a}\, e^{ap^2} \operatorname{erfc}(\sqrt{a}p)$
$te^{-t^2/(4a)} (a > 0)$	$2a\left[1 - 2\sqrt{a}pe^{ap^2} \operatorname{erfc}(\sqrt{a}p)\right]$
$\frac{e^{-2a\sqrt{t}}}{\sqrt{t}} (a > 0)$	$\frac{2}{\sqrt{p}} e^{a^2/p} \operatorname{erfc}\left(\frac{a}{\sqrt{p}}\right)$
$\frac{e^{-a^2/(4t)}}{\sqrt{t}} (a > 0)$	$\sqrt{\frac{\pi}{p}} e^{-a\sqrt{p}}$
$\frac{e^{-a^2/(4t)}}{t^{3/2}} (a > 0)$	$\frac{2\sqrt{\pi}\, e^{-a\sqrt{p}}}{a}$
$\sin \alpha t$	$\frac{\alpha}{p^2 + \alpha^2}$
$\cos \alpha t$	$\frac{p}{p^2 + \alpha^2}$

References

1. J. Crank, *The mathematics of diffusion*, 2nd edn. (Oxford University Press, Oxford, 1975)
2. H.S. Carslaw, J.C. Jaeger, *Conduction of heat in solids* (Oxford University Press, Oxford, 1959)
3. H. Bateman, The solution of a system of differential equations occurring in the theory of radioactive transformations. Proc. Cambridge Philos. Soc **15**(pt V), 423–427 (1910)
4. T.H. Pigford, P.L. Chambré, M. Albert, M. Harada, F. Iwamoto, T. Kanki, D. Leung, S. Masuda, S. Muraoka, D. Ting, *Migration of radionuclides through sorbing media: analytical solutions – II*, LBL-11616 (Lawrence Berkeley Laboratory, Berkeley, 1980)
5. K. Higashi, T.H. Pigford, Analytical models for migration of radionuclides in geologic sorbing media. J. Nucl. Sci. Technol. **17**, 700–709 (1980)

Chapter 10
Appendix C: Fundamentals of Chemical Equilibrium and Thermodynamics for Radioactive Waste Management

Shinya Nagasaki

10.1 Enthalpy and Entropy

10.1.1 Enthalpy

In thermodynamics, the total energy that the system under consideration has is called internal energy (U). Let w represent the amount of work done to a system and let q represent the amount of energy added to the system in the form of energy. Then, the amount of resultant change in internal energy, ΔU, can be calculated as Eq. (10.1).

$$\Delta U = q + w \tag{10.1}$$

This is the first law of thermodynamics. Let us consider here that if energy is given to a system in the form of heat or work, then $w > 0$, and, conversely, if energy is removed from the system in the form of heat or work, then $w < 0$.

Thermodynamics is often introduced in connection with a constant-volume process or a constant-pressure process. In this chapter, let us consider a constant-pressure process (for example, constant under atmospheric pressure). Now, let us consider the process in which a gas in a piston expands while the piston continues to be pressed by a constant external pressure p_{ex}. The work in this case can be expressed as

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$$w = -p_{\text{ex}} \int_{V_i}^{V_f} dV = -p_{\text{ex}}\left(V_f - V_i\right) \tag{10.2}$$

If, therefore, the change in volume is written as $\varDelta V = V_f - V_i$, then

$$w = -p_{\text{ex}} \varDelta V \tag{10.3}$$

If the pressure of the system is constant and the volume can change freely, enthalpy H can be defined as follows:

$$H = U + pV \tag{10.4}$$

where p is the pressure of the system and V is the volume of the system.

The enthalpy of a substance increases when the substance is heated. Heat capacity C_p (called constant-pressure heat capacity because it is heat capacity under constant pressure) corresponds to the temperature-dependent change in enthalpy. Hence,

$$C_p = \left(\frac{\partial H}{\partial T}\right)_p \tag{10.5}$$

If, therefore, heat capacity is constant in the temperature range of interest, then Eq. (10.6) holds true for temperature changes within the measurable range.

$$\varDelta H = C_p \varDelta T \tag{10.6}$$

This means that because enthalpy is equal to the amount of heat q added under constant pressure, Eq. (10.7) holds true.

$$q = C_p \varDelta T \tag{10.7}$$

Temperature-dependent changes in heat capacity may be negligibly small if the temperature range is narrow. If, however, temperature change must be taken into consideration, the empirical formula

$$C_{pm} = a + bT + \frac{c}{T^2} \tag{10.8}$$

is applied in some cases to the heat capacity per mole C_{pm} (called molar isobaric heat capacity) of the substance concerned. In the above equation, a, b and c are material-specific values that are not dependent on temperature. It is necessary here to keep in mind that when considering enthalpy change, it is common practice to consider changes in processes in which initial and final substances are in standard states (standard enthalpy change), $\varDelta H^\circ$. When reporting thermodynamic data, it is necessary to report data at 298.15 K (25.00 °C).

The standard enthalpy change due to a change in the physical state such as the phase transition from the solid phase to the liquid phase is called the standard transition enthalpy and expressed as $\varDelta_{\mathrm{trs}}H^{\circ}$. The standard reaction enthalpy $\varDelta_{\mathrm{r}}H^{\circ}$ due to chemical change (chemical reaction) such as

$$\mathrm{A} + 2\mathrm{B} \rightarrow 3\mathrm{C} + \mathrm{D}$$

can be evaluated by the equation

$$\varDelta_r H^{\circ} = \sum\nolimits_{\text{Production system}} \delta H_m^{\circ} - \sum\nolimits_{\text{Reaction system}} \delta H_m^{\circ} \tag{10.9}$$

where $H_m^{\circ}(i)$ is the standard molar enthalpy of chemical species i at the temperature of interest, and δ represents the stoichiometric number for the chemical equation, and the superscript m represents mole.

Another characteristic of the standard enthalpy is that the overall reaction enthalpy is the sum of the standard enthalpies of individual reactions.

These characteristics indicate that enthalpy at a particular temperature can be determined from the heat capacities and enthalpies at other temperatures. When a substance is heated from temperature T_1 to temperature T_2, if a phase transition does not occur between T_1 and T_2, then

$$H(T_2) = H(T_1) + \int_{T_1}^{T_2} C_p dT \tag{10.10}$$

and the relation

$$\varDelta_r H^{\circ}(T_2) = \varDelta_r H^{\circ}(T_1) + \int_{T_1}^{T_2} \varDelta_r C_p^{\circ} dT \tag{10.11}$$

holds true for the standard reaction enthalpy. In the above equation, $\varDelta_r C_p{}^{\circ}$ is evaluated as follows:

$$\Delta_r C_p^{\circ} = \sum\nolimits_{\text{Production system}} \delta C_{pm}^{\circ} - \sum\nolimits_{\text{Reaction system}} \delta C_{pm}^{\circ} \tag{10.12}$$

where the subscript m represents mole.

10.1.2 Entropy

Whether the chemical reaction of interest is a spontaneous process or a non-spontaneous process is shown by the second law of thermodynamics. The second law of thermodynamics incorporates entropy S, which is defined as

$$\Delta S_{\mathrm{tot}} > 0 \tag{10.13}$$

This means that the entropy of an independent system increases during a spontaneous change. In the above expression, S_{tot} is the total entropy of the system and its surroundings.

In thermodynamics, entropy is defined as

$$dS = \frac{dq_{\mathrm{rev}}}{T} \tag{10.14}$$

For a change of a measurable degree between two states i and f, we can write

$$\Delta S = \int_i^f \frac{dq_{\mathrm{rev}}}{T} \tag{10.15}$$

where q_{rev} is the energy given to the reversible path between the two states.

When a substance precipitates (solidifies) or boils (evaporates), the order (regularity) of the atoms constituting the substance changes. It is thought, therefore, that a phase transition is accompanied by a change in entropy. Because $q = \Delta_{trs}H$ under a constant pressure, the following relationship holds true.

$$\Delta_{\mathrm{trs}}S = \frac{\Delta_{\mathrm{trs}}H}{T_{\mathrm{trs}}} \tag{10.16}$$

For temperature-dependent changes in entropy, the following relationship holds true.

$$S(T_2) = S(T_1) + \int_{T_1}^{T_2} \frac{dq_{\mathrm{rev}}}{T} \tag{10.17}$$

Since the process of interest is a process under a constant pressure,

$$dq_{\mathrm{rev}} = C_p dT \tag{10.18}$$

Hence, under a constant pressure,

$$S(T_2) = S(T_1) + \int_{T_1}^{T_2} \frac{C_p dT}{T} \tag{10.19}$$

This section has dealt with fundamentals of enthalpy and entropy. Much of the thermodynamic data available for safety assessment of radioactive wastes, such as data on stability constants and solubility products associated with complex formation and surface complex formation constants for adsorption, are obtained experimentally at 1 atmosphere and 25 °C. For example, however, in the case of geological disposal at a depth greater than 300 m, it may be necessary to take into consideration the influence of the geothermal gradient of about 3 °C per 100 m, or in the case of burying heat-generating waste forms, it may be necessary to take account of the influence of heat generation. When dealing with thermodynamic data for safety assessment of such systems, it is necessary to identify the enthalpy and entropy of reaction and evaluate thermodynamic data under relevant temperature conditions.

10.2 Gibbs Energy and Chemical Equilibrium

10.2.1 Gibbs Energy

When considering a system that is in thermal equilibrium with its surroundings at temperature T, the concept of Gibbs energy G expressed as

$$G = H - TS \tag{10.20}$$

is introduced. The use of G makes it possible to obtain

$$dG_{T,\,p} < 0 \tag{10.21}$$

as a criterion for judging whether the process of interest changes spontaneously. This means that while temperature and pressure are constant, chemical reaction proceeds spontaneously so that Gibbs energy decreases. The above inequality is one of the most important conclusions drawn in the field of chemical thermodynamics. But why is it necessary to introduce the concept of G? An example suggesting the reason for that is the existence of spontaneous endothermic reactions. Since H increases in this type of reaction, the state of the system changes so that enthalpy increases and $dH > 0$. The spontaneity of reaction means that $dH > 0$ and $dG < 0$. This means that the entropy of the system increases to the extent of becoming greater than dH. It follows, therefore, that the endothermic reaction is driven by the increase in the entropy of the system, and the amount of such change is greater than is needed to make up for the decrease in the amount of entropy of the surroundings induced by the heat given to the system by the surroundings.

By using the standard reaction enthalpy and the standard reaction entropy, the standard reaction Gibbs energy $\Delta_r G^\circ$ can be defined:

$$\Delta_r G^\circ = \Delta_r H^\circ - T\Delta_r S^\circ \tag{10.22}$$

Similarly, the standard formation Gibbs energy $\Delta_f G^\circ$ can be defined as the standard reaction Gibbs energy needed to produce a compound from an element in the standard state. The standard reaction Gibbs energy, therefore, can be evaluated easily from the relationship

$$\Delta_r G^\circ = \sum\nolimits_{\text{Production system}} \delta \Delta_f G^\circ \sum\nolimits_{\text{Reaction system}} \delta \Delta_f G^\circ \tag{10.23}$$

10.2.2 Chemical Equilibrium

Chemical reaction proceeds until apparent change does not occur any longer even if both reactants and products coexist. Needless to say, in this state of chemical equilibrium, there may be cases where only products exist. In many systems, however, both reactants and products exist in a state of chemical equilibrium.

Now, let us consider a chemical equilibrium of the simplest kind, A $\leftrightarrows$ B. When a small amount $d\xi$ of A has changed into B, the amount of change of A is $dn_A = -d\xi$, and the amount of change of B is $dn_B = +d\xi$. The reaction Gibbs energy is defined as

$$\Delta_r G = \left(\frac{\partial G}{\partial \xi}\right)_{T,\ p} \tag{10.24}$$

And reaction does not proceed spontaneously when

$$\Delta_r G = 0 \tag{10.25}$$

By using this relationship, the composition of the reactants and products in the system in chemical equilibrium can be evaluated.

For information about chemical potential μ and activity α, refer to relevant textbooks. Now, let us consider the reaction

$$\mathrm{A} + 2\mathrm{B} \rightarrow 3\mathrm{C} + \mathrm{D}.$$

As the reaction proceeds by $d\xi$, reactants A and B and products C and D change, respectively, by the following amounts:

$$dn_A = -d\xi,\ \ dn_B = -2d\xi,\ \ dn_C = +3d\xi,\ \ dn_D = +d\xi.$$

As a result, the amount of small change in Gibbs energy under constant temperature and pressure conditions is

$$dG = (3\mu_C + \mu_D - \mu_A - 2\mu_B)d\xi \tag{10.26}$$

Hence,

$$\Delta_r G = \left(\frac{\partial G}{\partial \xi}\right)_{T,\ p} = -\mu_A - 2\mu_B + 3\mu_C + \mu_D \tag{10.27}$$

The chemical potential and activity of substance i are related as follows:

$$\mu_i = \mu_i^{\circ} + RT \ln \alpha_i \tag{10.28}$$

From this relationship, we obtain

$$\Delta_r G = \Delta_r G^\circ + RT\ln\frac{\alpha_C^3\alpha_D}{\alpha_A\alpha_B^2} \tag{10.29}$$

In a state of chemical equilibrium, $\Delta_r G = 0$. Since the activity in that condition has an equilibrium value, we have

$$K = \left(\frac{\alpha_C^3\alpha_D}{\alpha_A\alpha_B^2}\right)_{eq} \tag{10.30}$$

or, more generally,

$$K = \prod_i \alpha_i^{\delta_i} \tag{10.31}$$

The equilibrium constant K expressed in terms of activity is called the thermodynamic equilibrium constant. Then, we obtain

$$RT\ln K = -\Delta_r G^\circ \tag{10.32}$$

By differentiating the equation with respect to temperature, we obtain

$$\frac{d\ln K}{dT} = \frac{\Delta_r H^\circ}{RT^2} \tag{10.33}$$

This equation indicates that when the temperature of a system rises, the concentration of reactants increases in the case of an exothermic reaction, and the concentration of products increases in the case of an endothermic reaction.

The activity α introduced in Eq. (10.28) is a parameter that indicates the activity level (for example, reactivity) of substances in a real system. Under ideal conditions, activity equals concentration. For example, a reaction involving solutes in a high ionic strength aqueous solution differs from the reactivity of solutes of the same concentration in a low ionic strength aqueous solution. The difference between the activity α and the concentration c is called the activity coefficient γ, which is defined by $\alpha \equiv \gamma c$.

Equations (10.32) and (10.33) are exact equations. These make it possible, therefore, to estimate the equilibrium constant for the chemical reaction of interest from thermodynamic data and, therefore, estimate the composition (concentration) of the reactants and products in a state of chemical equilibrium. It is possible, by using these equations, to make accurate predictions of chemical reactions such as the concentration of radionuclide that will be dissolved in groundwater, concentration distributions of different chemical species, and the degree of adsorption.

10.3 Adsorption

Mechanisms by which atoms, molecules and ions are adsorbed onto the surfaces of substances can be broadly classified into two types. One is a mechanism called physical adsorption. Physical adsorption results from the van der Waals interaction between atoms, molecules or ions to be adsorbed (called adsorbates) and the substance that has the surface onto which they are adsorbed. Van der Waals interaction is long-range force, but its interaction force is small, and the amount of energy released when the adsorbate is physically adsorbed is roughly the same as the enthalpy of condensation, typically around 20 kJ/mol. The other mechanism is called chemical adsorption, in which the adsorbate is chemically bonded to the surface of the adsorbent. It is generally said that chemical adsorption tends to occur in areas where the coordination number is maximized. The enthalpy of chemical adsorption is greater than that of physical adsorption and is typically around 200 kJ/mol. The distance from the adsorbent surface to adsorbate atoms tends to be shorter in chemical adsorption than in physical adsorption.

The adsorption reaction is exothermic, and the fact that it is not endothermic is self-evident from Eq. (10.20). In the adsorption reaction, adsorbate particles moving in three-dimensional space are restrained on or near the adsorbent surface (two-dimensional). This means that a transition occurs from a high-entropy state to a low-entropy state. What "the occurrence of adsorption" means is that a state of equilibrium is reached as a result of adsorption, and a transition from a state of non-equilibrium to a state of equilibrium means a decrease in Gibbs energy. The reason is that if the amount of change in Gibbs energy, the change in enthalpy, and the change in entropy are represented by ΔG, ΔH, and ΔS, respectively, then the equation

$$\Delta G = \Delta H - T\Delta S \tag{10.33}$$

holds true, and ΔH must always be negative (exothermic) if $\Delta G < 0$ and $\Delta S < 0$.

10.3.1 Adsorption Isotherm Equation

Among the atoms, molecules and ions of interest, the atoms, molecules and ions staying dissolved in the groundwater without being adsorbed are thought to be in a state of dynamic equilibrium with the atoms, molecules and ions (adsorbate) adsorbed onto the rock (adsorbent) in contact with the groundwater. The ratio θ between the total number of effective adsorption sites on the adsorbent surface and the number of adsorption sites actually adsorbed by adsorbates is called the coverage ratio. The coverage ratio θ ranges between 0 and 1. A mathematical expression of changes in θ occurring depending on the atomic, molecular or ionic concentration at the temperature of the system of interest is called an adsorption isotherm equation.

The simplest and physically most realistic adsorption isotherm equation is based on the following assumptions.

(1) Adsorption does not occur beyond a monomolecular layer.
(2) The adsorbent surface is homogeneous, and all adsorption sites are equivalent.
(3) The ability of an adsorbate to be adsorbed to an adsorption site does not depend on the coverage ratio.

A state of equilibrium in this case is

$$\overline{\mathrm{S}}\ (\text{absorbent surface}) + \mathrm{M}\ (\text{atoms, molecules, ions}) \leftrightarrows \overline{\mathrm{SM}}$$

Let k_a and k_b represent the reaction rate constants for adsorption and desorption, respectively. The rate at which the coverage ratio changes because of adsorption is proportional to M (concentration C) and the number of unadsorbed adsorption sites, $N\ (1-\theta)$, where N is the total number of adsorption sites. The rate at which the coverage ratio changes because of desorption is proportional to the number of adsorbed species, $N\theta$.

$$\frac{d\theta}{dt} = k_a CN(1-\theta)\ \text{: absorption reaction} \tag{10.34}$$

$$\frac{d\theta}{dt} = -k_b N\theta\ \text{: desorption reaction} \tag{10.35}$$

In a state of equilibrium, the sum of these two reaction rates is 0. By solving the equations with respect to θ, we obtain the Langmuir adsorption isotherm.

$$\theta = \frac{KC}{1+KC} \tag{10.36}$$

$$K = \frac{k_a}{k_b} \tag{10.37}$$

Another adsorption isotherm equation is introduced here. Adsorption enthalpy often changes to smaller negative values as θ increases. This indicates that adsorption occurs preferentially at adsorption sites that are advantageous to the adsorbate–adsorbent combination in terms of energy. This is an example of failure of the aforementioned assumptions (2) and (3) for the Langmuir adsorption isotherm. The equation based on the assumption that adsorption enthalpy changes exponentially relative to concentration is called the Freundlich adsorption isotherm, which is expressed as

$$\theta = c_1 C^{1/c_2} \tag{10.38}$$

where c_1 and c_2 are constants.

Adsorption models based on these adsorption isotherm equations or adsorption mechanisms, combined with reaction rate theories, are used for mass transfer models such as the advection–dispersion model used for safety assessment of radioactive waste disposal in order to make rational and reliable safety assessment possible.

10.3.2 $\mathrm{K_d}$ *Model*

Radionuclides leaching into groundwater migrate while being adsorbed or desorbed by mechanisms of one kind or another involving buffers, rock, etc. Consequently, the rate of radionuclide migration is lower than the rate of groundwater movement. In safety assessment or performance assessment of radioactive waste disposal, it is common practice to simplistically regard sorption and desorption reaction as distribution between water and solid (buffer or rock in this case) surfaces; assume that partition equilibrium occurs instantaneously and reversibly and adsorption does not reach saturation; and evaluate the barrier function of buffers and rock by using the sorption distribution coefficient K_d as an indicator of equilibrium (Sect. 7.2.2). This approach is called the K_d model.

While radionuclides migrate in groundwater, their migration is likely to be retarded by various sorption phenomena. In view of the complexity and heterogeneity of geological formations, however, it is difficult to interpret individual phenomena and evaluate their retardation effect.

The two adsorption mechanisms mentioned above—physical adsorption and chemical adsorption—are further classified into three types:

Mechanism (a), surface complexation: coordinate bond formation between atoms or atom groups at a solid surface and an adsorbate.

Mechanism (b), electric interaction: the distribution of ions near a solid surface due to the influence of electric charge (the incorporation of ions into the electric double layer).

Mechanism (c), physical adsorption.

Let $\overline{\mathrm{S}}$ represent an adsorption site. In Mechanism (a) or (c), the adsorption reaction in the case where there is no secondary reaction involving the adsorbate can be expressed as

$$\overline{\mathrm{S}} + \mathrm{M} \leftrightarrows \overline{\mathrm{SM}} \tag{10.39}$$

In Mechanism (b), it can be expressed as

$$\overline{\mathrm{S\cdots A}} + \mathrm{M} \leftrightarrows \overline{\mathrm{S\cdots M}} + \mathrm{A} \tag{10.40}$$

An overbar indicates proximity to a surface. The dotted lines in Eq. (10.40) indicate a state of being bound by Coulomb force. Now, if it is assumed that changes in Gibbs energy due to these reactions are constant regardless of the progress of reaction, we obtain the relationship

$$K = \exp\left(-\frac{\varDelta G^{\circ}}{RT}\right) = \frac{\overline{[\mathrm{SM}]}}{\overline{[\mathrm{S}]}\,[\mathrm{M}]} \tag{10.41}$$

$$\overline{[\mathrm{S}]}_{\mathrm{T}} = \overline{[\mathrm{SM}]} + \overline{[\mathrm{S}]} \tag{10.42}$$

for Eq. (10.39) and the relationship

$$K = \exp\left(-\frac{\varDelta G^{\circ}}{RT}\right) = \frac{\overline{[\mathrm{S\cdots M}]}\,[\mathrm{A}]}{[\mathrm{S\cdots A}][\mathrm{M}]} \tag{10.43}$$

$$\overline{[\mathrm{S}]}_{\mathrm{T}} = \overline{[\mathrm{S\cdots M}]} + \overline{[\mathrm{S\cdots A}]} \tag{10.44}$$

for Eq. (10.40). In the above equations, an overbar indicates the adsorption site or adsorbate density in the near-surface region, and $\overline{[\mathrm{S}]}_{\mathrm{T}}$ represents the total adsorption per unit area in the near-surface region. These equations are based on the assumption that the activity of the adsorbate in the near-surface region is proportional to the density of adsorbate in that region. The adsorption expressed by Eq. (10.41) is Langmuir adsorption expressed by the Langmuir adsorption isotherm mentioned earlier. The adsorption with a normal distribution of $\varDelta G^{\circ}$ (in other words, with a logarithmic normal distribution of K) is Freundlich adsorption. Even in such cases, $\varDelta G^{\circ}$ can be deemed approximately to be constant if the degree of progress is within a certain range and therefore can be expressed by Eq. (10.41) or (10.43). In such cases, the ratio between the density of the adsorbate of interest in the near-surface region and the concentration in the solution can be expressed as

$$K_d^{*} = \frac{\overline{[\mathrm{SM}]}}{[\mathrm{M}]} = K\,\overline{[\mathrm{S}]} = K\left(\overline{[\mathrm{S}]}_{\mathrm{T}} - \overline{[\mathrm{SM}]}\right) \tag{10.45}$$

or

$$K_d^{*} = \frac{\overline{[\mathrm{S\cdots M}]}}{[\mathrm{M}]} = K\frac{\overline{[\mathrm{S\cdots M}]}}{[\mathrm{A}]} = K\frac{\left(\overline{[\mathrm{S}]}_{\mathrm{T}} - \overline{[\mathrm{SM}]}\right)}{[\mathrm{A}]} \tag{10.46}$$

Since $\overline{[\mathrm{S}]}_{\mathrm{T}}$ and [A] change depending on pH and other conditions, and $\overline{[\mathrm{S}]}$ and $\overline{[\mathrm{S\cdots A}]}$ change depending on the degree of progress of adsorption reaction, K_d* is not a constant. If, however, the solution conditions remain unchanged and only a very small amount of adsorbate is adsorbed at an excessively large number of adsorption sites, $\left[\overline{\text{S}}\right]$ and $\left[\overline{\text{S}\cdots\text{A}}\right]$ are roughly equal to $[\text{S}]_\text{T}$, and [A] is more or less constant. The constant K_d*, therefore, may be thought of as nearly constant depending on conditions. In this case, K_d* indicates the adsorbate distribution equilibrium between the solution and the surface, which is the same relationship given by Henry's law expressing the relationship between the amount of gas dissolved in liquid and pressure. This type of adsorption, therefore, is called Henry adsorption, and K_d* in this case is called the adsorption distribution coefficient. The concept of K_d* includes the following assumptions.

(1) The Gibbs energy of adsorption is constant.
(2) Adsorption sites are in excessively large supply, and there is no influence of the saturation of adsorption sites.
(3) The solution conditions at the time of adsorption are constant.
(4) In the adsorption reaction, Mechanism (a), (b) or (c) is dominant.

It is difficult to determine the area of the surface or the volume of the near-surface region where adsorption occurs. It may be thought, however, that in the case of homogeneous solid having constant physical properties (e.g., solid particle size, fine crack size and distribution, net-like or laminar hydrophilic voids), the area of the surface or the volume of the near-surface region accessible by the solution is proportional to the mass of the solid. To consider the adsorption reaction, therefore, the apparent concentration obtained by dividing the amount of adsorbate accumulated in the near-surface region, in place of the density of the adsorbate in the near-surface region, by the mass of the solid is used, and it is referred to as the solid phase concentration:

$$\text{Solid phase concentrations } S = \frac{\text{Amount of adsorbate accumulated in near-surface region [mol]}}{\text{Mass of the solid providing the surface [kg]}}. \tag{10.47}$$

This apparent concentration S, which is thought to be proportional to $\left[\overline{\text{S}}\right]$ and $\left[\overline{\text{S}\cdots\text{A}}\right]$, is used in place of the activity at the surface. The concentration C of the adsorbate of interest in the solution is proportional to the concentration of the adsorbate if the solution conditions are constant. Therefore, the distribution ratio of the adsorbate of interest

$$K_d\left(\text{m}^3/\text{kg}\right) = \frac{S(\text{mol/kg})}{C(\text{mol/m}^3)} \tag{10.48}$$

is proportional to the K_d* in Eqs. (10.45) and (10.46) and may be regarded as a coefficient that is constant under certain conditions. This concept of K_d includes not only the assumptions (1)–(4) mentioned above but also two more assumptions.

(5) Physical and chemical properties of the surface and the near-surface region do not change with time.
(6) There is a relationship of proportionality between the area of the surface or the volume of the near-surface region and the mass of the solid, and its ratio is constant.

Thus, the sorption distribution coefficient, derived by modifying K_d defined with respect only to adsorption phenomena so as to allow for an absorption-related distribution coefficient, is used for safety assessment or performance assessment of radioactive waste disposal. It can be used, for example, by applying it to a one-dimensional advection–diffusion model

$$R\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - u\frac{\partial C}{\partial x} \tag{10.49}$$

$$R = 1 + \frac{1-\varepsilon}{\varepsilon}\rho K_d \tag{10.50}$$

to consider radionuclide migration through a porous formation having the porosity ε for pores that can be involved in the transport of substances and the solid density ρ. In the equation, D is the dispersion coefficient; u is the flow velocity of the medium (for example, groundwater); C is radionuclide concentration; and R, called the retardation factor, is the ratio of the rate of movement of medium to the rate of movement of substance of interest and indicates how slow the substance moves relative to the groundwater. The use of K_d in connection with the movement of substances has been discussed in Chap. 7.

10.4 Exercises with Solutions

Some thermodynamics- and chemical equilibrium-related exercises with solutions are shown below.

[Exercise 1]

Calculate the entropy change occurring when 1 mol of water is heated from −50 °C to 500 °C at 1 atm.

Heat of fusion of ice at 0 °C and 1 atm: 6,008 J/mol
Heat of evaporation of water at 100 °C and 1 atm: 40,660 J/mol
Molar isobaric heat capacity of ice: 35.56 J/(K·mol)
Molar isobaric heat capacity of water: 75.31 J/(K·mol)
Molar isobaric heat capacity of steam: $(30.20 + 0.00992\,T)$ J/(K·mol)

(Example of Solution)

The process from −50 °C to 500 °C can be divided into five stages:

H_2O (solid, −50 °C)
↓ Stage I

H_2O (solid, 0 °C)
↓ Stage II
H_2O (liquid, 0 °C)
↓ Stage III
H_2O (liquid, 100 °C)
↓ Stage IV
H_2O (gas, 100 °C)
↓ Stage V
H_2O (gas, 500 °C)

At Stages I, III and V, entropy increases because of heating of a phase.

$$\Delta S = \int_{T_1}^{T_2} \frac{C_p}{T} dT = \int_{T_1}^{T_2} C_p d\ln T$$

At Stages II and IV, entropy increases as a result of a phase transition.

$$\Delta S_{\text{trans}} = \frac{\Delta H_{\text{trans}}}{T_{\text{trans}}}$$

The entropy change can be calculated as follows.

$$\begin{aligned}\Delta S &= \Delta S_{\text{I}} + \Delta S_{\text{II}} + \Delta S_{\text{III}} + \Delta S_{\text{IV}} + \Delta S_{\text{V}} \\ &= \left(35.56\ln\frac{273}{223} + \frac{6\,008}{273} + 75.31\ln\frac{673}{273} + \frac{40\,660}{373} + \int_{373}^{773} \frac{30.20 + 0.00992T}{T} dT\right) \\ &= 187.7(\text{J/K} \cdot \text{mol})\end{aligned}$$

[Exercise 2]
The standard formation enthalpy for gaseous H_2O is −241.82 kJ/mol at 298 K. Calculate this value at 100 °C by using the following molar isobaric heat capacity values: 33.58 J/(K·mol) for H_2O (g), 28.84 J/(K·mol) for H_2 (g), and 29.37 J/(K·mol) for O_2 (g). Assume that heat capacity is not dependent on temperature.

(Example of Solution)
It is assumed that no substance undergoes a phase transition in the temperature range of interest. When a substance is heated from T_1 to T_2, its enthalpy becomes $H(T_2) = H(T_1) + \int_{T_1}^{T_2} C_p dT$. Since heat capacity C_p is not dependent on temperature, the integration term is simply the product of the temperature difference $(T_1 - T_2)$ and the heat capacity C_p.

The reaction is $H_2(g) + \frac{1}{2}O_2(g) \rightarrow H_2O(g)$. Therefore,

$$C_p = C_p(H_2O, g) - \left\{C_p(H_2, g) + \frac{1}{2}C_p(O_2, g)\right\} = -9.94 \ \text{J/(K} \cdot \text{mol)}$$

Hence,

$$\Delta H(373\text{K}) = -241.82\ \text{kJ/mol} + (75\text{K}) \cdot (-9.94\ \text{J/K} \cdot \text{mol}) = -242.6\ \text{kJ/mol}.$$

The temperature dependence of entropy shown in Exercises 1 and 2 is used to evaluate the temperature dependence of thermodynamic data (equilibrium constants and Gibbs energy) obtained under the standard conditions and integrated into a database. Conversely, if the entropy and enthalpy of the system under consideration have been evaluated in advance, the results can be used as indices for evaluating thermodynamic data or selecting the most reliable data from variously proposed thermodynamic data.

[Exercise 3]
Dissolution equilibrium of AgCl (s) in a solution can be described as follows.

$$\begin{aligned} &\text{AgCl(s)} \leftrightarrows \text{Ag}^+ + \text{Cl}^- \\ &\log K = \log \frac{[\text{Ag}^+][\text{Cl}^-]}{[\text{AgCl(s)}]} = -9.42 \end{aligned}$$

If the solid phase AgCl (s) is present in a large quantity, its concentration [AgCl (s)] can be deemed to be equal to 1. Therefore,

$$\log K_{sp} = \log[\text{Ag}^+][\text{Cl}^-] = -9.42$$

Find the Cl^- concentration that meets this constraint under the following conditions.

(i) In pure water
(ii) 0.01 mol/L AgNO_3 solution

(Example of Solution)

(i) In pure water, Ag^+ ions and Cl^- ions are supplied solely by AgCl (s). Consequently, $[\text{Ag}^+] = [\text{Cl}^-]$ always holds true.

$$\begin{aligned} &\therefore \log K_{sp} = \log[\text{Cl}^-]^2 = -9.42 \\ &\therefore \log[\text{Cl}^-] = -\frac{9.42}{2} = -4.71 \end{aligned}$$

Hence, $[\text{Cl}^-] = 10^{-4.71} = 1.95 \times 10^{-5}$ mol/l.

(ii) AgNO_3 salts dissociate completely in a solution. Hence, $[\text{Ag}^+] = 10^{-2}$ mol/L.

$$\begin{aligned} &\therefore \quad \log K_{sp} = \log[\text{Cl}^-] + \log[\text{Ag}^+] = \log[\text{Cl}^-] - 2 \\ &\therefore \quad \log[\text{Cl}^-] = -9.42 + 2 = -7.42 \\ &\therefore \quad [\text{Cl}^-] = 10^{-7.42} = 3.80 \times 10^{-8}\text{mol/l} \end{aligned}$$

Because of the contribution of the common ions (Ag^+), the concentration of $[\text{Cl}^-]$ decreases.

Imagine an experiment in which common table salt is added to a beaker of water. At first, the salt dissolves in the water. As more and more salt is added, however, salt gradually becomes insoluble and accumulates at the bottom of the beaker. This result indicates that there is a limit to the solubility of common table salt, and it does not dissolve in water beyond that solubility limit. We can deduce that this result is applicable not only to common table salt but also to any other substance.

This means that no matter how much of a radionuclide is contained in radioactive waste, the radionuclide does not dissolve in groundwater beyond the solubility limit of each solid form. When considered simplistically, the radionuclide present at a concentration at or below the solubility limit in the vicinity of a solidified waste form is transported through the surrounding engineered barrier or natural barrier by advection and diffusion under the boundary conditions determined by the upper limit of concentration. In performance assessment or safety assessment of a radioactive waste disposal system, therefore, it is important to understand in what solid forms radionuclides are present and know what the solubility product (thermodynamic data indicating solubility) values are. In this example exercise, the solubility of a substance has been calculated by using the solubility product, and the influence of common ions present in groundwater has been considered.

[Exercise 4]

Thermodynamic equilibrium constants for protonation, $\log\beta_x{}^{\circ}$, for ethylenediaminetetraacetic acid (EDTA) are given as follows.

$$\log\beta_1^{\circ} = \log\frac{\alpha_{\left(\mathrm{HL^{3-}}\right)}}{\alpha_{\left(\mathrm{L^{4-}}\right)}\alpha_{(\mathrm{H^+})}} = 10.948, \quad \log\beta_2^{\circ} = \log\frac{\alpha_{\left(\mathrm{H_2L^{2-}}\right)}}{\alpha_{\left(\mathrm{L^{4-}}\right)}\alpha^2_{(\mathrm{H^+})}} = 17.221,$$

$$\log\beta_3^{\circ} = \log\frac{\alpha_{(\mathrm{H_3L^-})}}{\alpha_{\left(\mathrm{L^{4-}}\right)}\alpha^3{}_{(\mathrm{H^+})}} = 20.359, \quad \log\beta_4^{\circ} = \log\frac{\alpha_{(\mathrm{H_4L})}}{\alpha_{\left(\mathrm{L^{4-}}\right)}\alpha^4_{(\mathrm{H^+})}} = 22.583$$

Given these, calculate the consecutive acid dissociation constants (concentration equilibrium constants)

$$pK\alpha_1 = -\log\frac{[\mathrm{H_3L^-}][\mathrm{H^+}]}{[\mathrm{H_4L}]}, \quad pK\alpha_2 = -\log\frac{\left[\mathrm{H_2L^{2-}}\right][\mathrm{H^+}]}{[\mathrm{H_3L^-}]},$$

$$pK\alpha_3 = -\log\frac{\left[\mathrm{HL^{3-}}\right][\mathrm{H^+}]}{\left[\mathrm{H_2L^{2-}}\right]}, \quad pK\alpha_4 = -\log\frac{\left[\mathrm{L^{4-}}\right][\mathrm{H^+}]}{\left[\mathrm{HL^{3-}}\right]}$$

at an ionic strength I of 0.1 mol/L. In the above equations, α_i is the activity of chemical species i, where α_i is given as the product of the concentration of i $[i]$ and the activity coefficient γ_i. Calculate the activity coefficient $\log\gamma_i$ by using the Davies model, which is expressed as

$$\log\gamma_i = -\frac{0.51 z_i^2 \sqrt{I}}{1+\sqrt{I}} + 0.15 z_i^2 I$$

where I is the ionic strength and z_i is the electric charge of chemical species i.

The letter "*p*" placed before a constant, such as that in $pK_{\alpha 1}$, is a calculation operator used only in chemistry representing "−log." The "pH" shown earlier in this book is formed by this "*p*" and [H+], representing hydrogen ion concentration. Strictly speaking, therefore, the correct form is *p*H instead of pH.

(Example of Solution)

The equilibrium constant $\log\beta_x$ at $I = 0.1$ mol/L is calculated from the thermodynamic equilibrium constant for protonation, $\log\beta_x{}^\circ$:

$$\begin{aligned}
\log\beta_1^\circ &= \log\frac{\alpha_{(\mathrm{HL}^{3-})}}{\alpha_{(\mathrm{L}^{4-})}\alpha_{(\mathrm{H}^+)}} = \log\frac{\gamma_{\mathrm{HL}^{3-}}\left[\mathrm{HL}^{3-}\right]}{\gamma_{\mathrm{L}^{4-}}\left[\mathrm{L}^{4-}\right]\gamma_{\mathrm{H}^+}\left[\mathrm{H}^+\right]} \\
&= \log\beta_1 + \log\gamma_{\mathrm{HL}^{3-}} - \log\gamma_{\mathrm{L}^{4-}} - \log\gamma_{\mathrm{H}^+} \\
\log\beta_1 &= \log\beta_1^\circ - \log\gamma_{\mathrm{HL}^{3-}} + \log\gamma_{\mathrm{L}^{4-}} + \log\gamma_{\mathrm{H}^+} \\
&= 10.948 - (-0.968) - 1.721 - 0.108 = 10.087
\end{aligned}$$

Similarly, we obtain

$$\begin{aligned}
\log\beta_2 &= \log\beta_2^\circ - \log\gamma_{\mathrm{H_2L}^{2-}} + \log\gamma_{\mathrm{L}^{4-}} + 2\log\gamma_{\mathrm{H}^+} \\
&= 17.221 - (-0.430) - 1.721 - 2\cdot 0.108 = 15.714 \\
\log\beta_3 &= \log\beta_3^\circ - \log\gamma_{\mathrm{H_3L}^{-}} + \log\gamma_{\mathrm{L}^{4-}} + 3\log\gamma_{\mathrm{H}^+} \\
&= 20.359 - (-0.108) - 1.721 - 3\cdot 0.108 = 18.422 \\
\log\beta_4 &= \log\beta_4^\circ - \log\gamma_{\mathrm{H_4L}} + \log\gamma_{\mathrm{L}^{4-}} + 4\log\gamma_{\mathrm{H}^+} \\
&= 22.583 - 0 - 1.721 - 4\cdot 0.108 = 20.430
\end{aligned}$$

Hence,

$$pK_{\alpha_1} = -\log\frac{[\mathrm{H_3L^-}][\mathrm{H^+}]}{[\mathrm{H_4L}]} = \log\frac{\left[\mathrm{L}^{4-}\right][\mathrm{H}^+]^4}{[\mathrm{H_4L}]}\frac{[\mathrm{H_3L^-}]}{\left[\mathrm{L}^{4-}\right][\mathrm{H}^+]^3} = \log\beta_4 - \log\beta_3$$

$$= 2.008$$

$$pK_{\alpha_2} = -\log\frac{\left[\mathrm{H_2L}^{2-}\right][\mathrm{H^+}]}{[\mathrm{H_3L^-}]} = \log\frac{\left[\mathrm{L}^{4-}\right][\mathrm{H}^+]^3}{[\mathrm{H_3L^-}]}\frac{\left[\mathrm{H_2L}^{2-}\right]}{\left[\mathrm{L}^{4-}\right][\mathrm{H}^+]^2} = \log\beta_3 - \log\beta_2$$

$$= 2.708$$

$$pK_{\alpha_3} = -\log\frac{\left[\mathrm{HL}^{3-}\right][\mathrm{H^+}]}{[\mathrm{H_2L}]} = -\log\frac{\left[\mathrm{L}^{4-}\right][\mathrm{H}^+]^2}{\left[\mathrm{H_2L}^{2-}\right]}\frac{\left[\mathrm{HL}^{3-}\right]}{\left[\mathrm{L}^{4-}\right][\mathrm{H}^+]} = \log\beta_2 - \log\beta_1$$

$$= 5.627$$

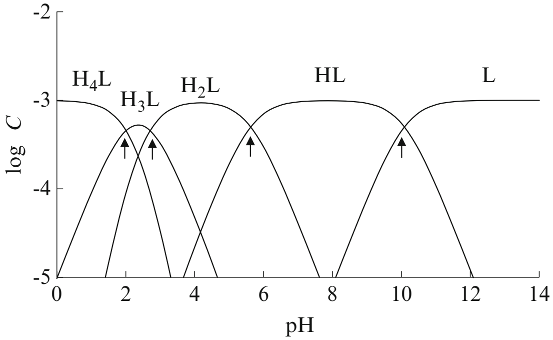

Fig. 10.1 Distribution of EDTA species as a function of pH at total EDTA concentration of 1×10^{-3} mol/L

$$pK_{\alpha_4} = -\log\frac{[\mathrm{L}^{4-}][\mathrm{H}^+]}{[\mathrm{HL}^{3-}]} = \log\beta_1 = 10.087$$

The pH dependence of EDTA chemical species in the case where the total concentration of EDTA is assumed to be 1×10^{-3} mol/L is shown in Fig. 10.1 (the minus charge symbols are omitted). It can be seen that a pK_{a_x} value is a pH value corresponding to a point where the concentrations of adjacent chemical species are equal. For example, the most dominant chemical species at pH = 6 is HEDTA^{3-} followed by $\mathrm{H_2EDTA}^{2-}$ (Fig. 10.1).

Nuclides that must be taken into consideration in connection with radioactive waste disposal include ones whose most stable chemical forms (speciation) change depending on the pH of groundwater. For example, in an oxygen-saturated water solution that does not contain hydrogen carbonate ions, ^{237}Np (neptunium), which is an alpha-emitting nuclide with a half life of 2.14 million years, changes (hydrolyzes) to $\mathrm{NpO_2}^+$, $\mathrm{NpO_2OH}^0$ and then $\mathrm{NpO_2(OH)_2}^-$ as pH increases. Since rock surfaces are often charged negatively upon contact with groundwater, it can be simply stated that $\mathrm{NpO_2}^+$ ions are adsorbed but the adsorption of $\mathrm{NpO_2(OH)_2}^-$ ions is not equally promoted. For the identification of speciation, thermodynamic data including acid dissociation constants for molecules that can become ligands such as EDTA, and complex stability constants for nuclides are important.

[Exercise 5]

Draw a log C–pe graph showing the pe dependence of uranium species at an ionic strength I of 1.0 mol/L, $[\mathrm{U}]_{\mathrm{total}} = 1$ mmol/L and an acidity of 1 mol/L (pH = 0) when $-15 \leq \mathrm{pe} \leq 15$. The value of pe at 25 °C is given by $\mathrm{pe} = E_h \times \frac{F}{RT\ln(10)} = \frac{E_h(\mathrm{mV})}{59.2}$. Consider the reactions shown below, for which equilibrium constants are given (concentration equilibrium constant: $I = 1.0$ mol/L). In the first three reactions, H_2O is omitted.

$$4H^+ + 2e^- + UO_2^{2+} \leftrightarrows U^{4+} \qquad : \log K_1 = 11.1$$

$$4H^+ + 3e^- + UO_2^{2+} \leftrightarrows U^{3+} \qquad : \log K_2 = 0.14$$

$$3H^+ + 2e^- + UO_2^{2+} \leftrightarrows U(OH)^{3+} \qquad : \log K_3 = 9.13$$

$$e^- + UO_2^{2+} \leftrightarrows UO_2^{+} \qquad : \log K_4 = 0.794$$

$$-H^+ + (H_2O) \leftrightarrows OH^- \qquad : \log K_5 = -13.65$$

(Example of Solution)

Besides the five equilibrium constant formulae shown above, in the system of interest, material balance is kept in terms of hydrogen ion concentration and uranium ion concentration. With the given condition pH = 0, hydrogen ion concentration $[H^+]$ is constant at $[H^+] = 1$ mol/L. From a given condition, total uranium concentration is also constant at $[U]_{total} = 1$ mmol/L. Hence, the following equation holds true:

$$[U]_{total} = \left[UO_2{}^{2+}\right] + \left[U^{4+}\right] + \left[U^{3+}\right] + \left[U(OH)^{3+}\right] + \left[UO_2{}^{+}\right] = 1 \text{ mmol/l.}$$

By using the definition of the equilibrium constant, this equation can be rewritten in terms of $[H^+]$, $[e^-]$ and $[UO_2{}^{2+}]$ (hereinafter shown as [M]) as follows:

$$\begin{aligned}[U]_{total} = {} & [M] + K_1[H^+]^4[e^-]^2[M] + K_2[H^+]^4[e^-]^3[M] + K_3[H^+]^3[e^-]^2[M] \\ & + K_4[e^-][M]\end{aligned}$$

Since $[H^+] = 1$ mol/L, we obtain

$$[U]_{total} = [M] + (K_1 + K_3)[e^-]^2[M] + K_2[e^-]^3[M] + K_4[e^-][M].$$

Solving this with respect to [M] gives

$$[M] = \frac{[U]_{total}}{1 + (K_1 + K_3)[e^-]^2 + K_2[e^-]^3 + K_4[e^-]}.$$

By using the above formula, [M] (i.e., $[UO_2{}^{2+}]$) at each pe point ($[e] = 10^{-pe}$) can be calculated. Also, by using $[e^-]$, $[UO_2{}^{2+}]$ and the equilibrium constant given in the exercise, the concentration of all uranium species other than $[UO_2{}^{2+}]$ can be calculated. The results are shown below (Fig. 10.2).

[Exercise 6]

Pure water is composed of a very small amount of hydrogen ions (H^+), a very small amount of hydroxide ions (OH^-) and a large amount of water (H_2O). At 25 °C, the following equilibrium holds:

(1) $H^+ + OH^- = H_2O : \log K_w = -14.0$

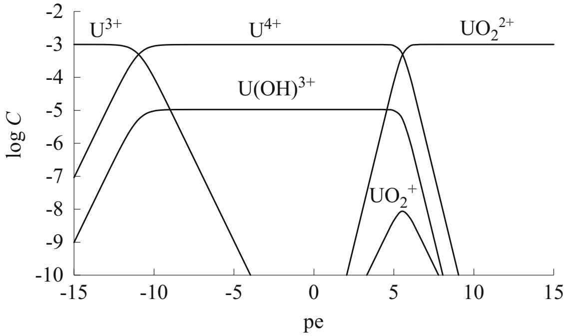

Fig. 10.2 Distribution of U(IV, V, VI) as a function of pe. Total U concentration is 1 mmol/l and pH = 0

From this equilibrium reaction and the electrical neutrality condition ($[H^+] = [OH^-]$) between all ions (i.e., H^+ and OH^-), the pH of pure water can be calculated as $pH = -\log[H^+] = 7.0$. The pH, however, of water that is open to the air and is in equilibrium with the atmospheric pressure is not necessarily 7.0. That is because of the very small soluble portions of the gases contained in the air. The most influential gas is carbon dioxide (CO_2).

In water, carbon dioxide reacts with water molecules and exists as carbonic acid (H_2CO_3), hydrogen carbonate ions (HCO_3^-) or carbonate ions (CO_3^{2-}), and the ratios of those components vary depending on pH. The chemical reactions expressing such water-related equilibrium are reaction (1) shown above and reactions (2)–(4) shown below.

(2) $H^+ + CO_3^{2-} = HCO_3^- : \log K_1 = 9.9$

(3) $H^+ + HCO_3^- = H_2CO_3 : \log K_2 = 6.1$

(4) $CO_2^{(g)} + H_2O = H_2CO_3 : \log K_3 = -1.5$

Concerning this system, answer the following questions:

(i) Express the equilibrium constants K_w, K_1, K_2 and K_3 for reactions (1)–(4) in terms of the concentration of chemical species (H^+, OH^-, H_2CO_3, HCO_3^-, CO_3^{2-}) and the partial pressure p of CO_2 contained in the air. Assume that as a commonly used value, that the concentration of water $[H_2O]$ is 1.0.

(ii) According to a World Data Centre for Greenhouse Gases analysis, the global average of carbon dioxide concentration in 2006 was 381 ppm. Assuming an atmospheric pressure of 1.0 atm, calculate the partial pressure p of carbon dioxide.

(iii) Draw a graph of the concentration of each chemical species ($[H^+]$, $[OH^-]$, $[H_2CO_3^-]$, $[HCO_3^-]$, $[CO_3^{2-}]$) as a function of pH. Assume a pH range of 2–10, and use a logarithmic scale for the vertical axis.

(iv) In the solution, electric charge balance is conserved among [H+], $[HCO_3^-]$ and $[CO_3^{2-}]$ ions. Hence, the equation

$$[\mathrm{H}^+] = [\mathrm{HCO_3}^-] + 2\left[\mathrm{CO_3}^{2-}\right] + [\mathrm{OH}^-]$$

holds. By simultaneously solving this equation and the equations derived in (i), hydrogen ion concentration can be calculated. From this, calculate pH. (Although it is not impossible to simultaneously solve the equations purely mathematically, a close look at the graph drawn in (iii) reveals that it is highly likely that the equations can be simplified. Use a logarithmic scale for the vertical axis.)

(v) It is generally said that the average carbon dioxide concentration before the Industrial Revolution was 280 ppm. What is the pH of the water that was in equilibrium with the air in those days? What will it be like if the concentration increases to 500 ppm in the future?

(vi) If CO_2 is blown into pure water at 1 atm, what will the pH be?

(Example of Solution)

(i)

$$K_w = [\mathrm{H}^+][\mathrm{OH}^-], \quad K_1 = \frac{[\mathrm{HCO_3}^-]}{[\mathrm{H}^+]\left[\mathrm{CO_3}^{2-}\right]}, \quad K_2 = \frac{[\mathrm{H_2CO_3}]}{[\mathrm{H}^+][\mathrm{HCO_3}^-]},$$

$$K_3 = \frac{[\mathrm{H_2CO_3}]}{p}$$

(ii) Because the concentration is 381 ppm at 1.0 atm, the partial pressure is $p = 3.81 \times 10^{-4}$atm $= 10^{-3.4}$atm.

(iii) By using the equilibrium constant values given for reactions (1) through (4), the relations derived in (i) above and $\mathrm{pH} = -\log[\mathrm{H}^+]$, the concentration of each chemical species can be expressed as a function of pH:

$$[\mathrm{H}^+] = 10^{-\mathrm{pH}}$$

$$[\mathrm{OH}^-] = \frac{K_w}{[\mathrm{H}^+]} = 10^{\mathrm{pH}-14.0}$$

$$[\mathrm{H_2CO_3}] = pK_3 = 10^{-3.4} \cdot 10^{-1.5} = 10^{-4.9}$$

$$\left[\mathrm{HCO_3^-}\right] = \frac{[\mathrm{H_2CO_3}]}{[\mathrm{H}^+]K_2} = \frac{10^{-4.9}}{10^{-\mathrm{pH}} \cdot 10^{6.1}} = 10^{\mathrm{pH}-11.0}$$

$$\left[\mathrm{CO_3}^{2-}\right] = \frac{[\mathrm{HCO_3}^-]}{[\mathrm{H}^+]K_1} = \frac{10^{\mathrm{pH}-11.0}}{10^{-\mathrm{pH}} \cdot 10^{9.9}} = 10^{2\mathrm{pH}-20.9}$$

By plotting these, we obtain the following diagram (Fig. 10.3):

(iv) The pH value to be determined can be calculated by substituting the equations obtained in (iii) into the electrical neutrality condition equation $[\mathrm{H}^+] = [\mathrm{HCO_3}^-] + 2[\mathrm{CO_3}^{2-}] + [\mathrm{OH}^-]$. Examination of the quantities shown on the right hand side while looking at the graph obtained in (iii) reveals

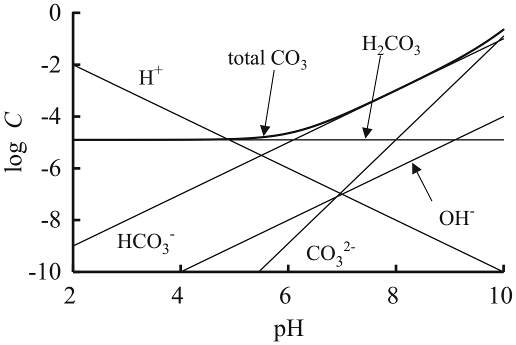

Fig. 10.3 Distribution of H^+, OH^-, H_2CO_3, HCO_3^- and CO_3^{2-} as a function of pH

that $[HCO_3^-]$ is dominant in terms of anion concentration, followed by $[OH^-]$ at $pH < 7$ and $[CO_3^{2-}]$ at $pH > 7$.

The first step, therefore, is to approximate the electrical neutrality condition as $[H^+] = [HCO_3^-]$. By substituting the formulae of (iii), we obtain $10^{-pH} = 10^{pH-11.0}$. Hence, pH = 5.5. Now that it has been found that the pH value is less than 7, we pay attention to $[OH^-]$, too, and we find that $10^{pH} = 10^{pH-11.0} + 10^{pH-14.0}$ holds. By solving this quadratic equation, we obtain pH = 5.4998 and find that the approximation obtained at first, namely, pH = 5.5, is sufficiently accurate.

(v) By repeating the calculations of (iii) and (iv) above assuming $p = 2.80 \times 10^{-4}$ atm $= 10^{-3.55}$ atm, we obtain pH = 5.6. At 500 ppm, $p = 5.00 \times 10^{-4}$ atm $= 10^{-3.3}$ atm. Hence, pH = 5.45.

(vi) pH = 3.8

The example shown here deals only with the influence of carbon dioxide. The pH of groundwater and other natural water, however, is determined as a result of equilibrium involving the components of the rock in contact with the water, influent water and the air. Because adsorption to precipitates and suspended solids is also involved, there are cases where, depending on the time scale under consideration, explanation cannot be made by applying thermodynamics alone. It is very difficult to describe the chemistry of natural water, but we can begin by discussing chemical equilibrium by focusing on main components. One of the findings from the example shown above is that, as determined above, as long as water is in contact with the air, the pH of the water is 7 or less, indicating that the water is weakly acidic. The pH of water, therefore, is 7 or less even without the influence of acid rain.

Answers to Exercises

Chapter 2

1. (1) Let the generated amount of depleted uranium be W and the required amount of natural uranium be N. The material balance of U and the material balance of ^{235}U provide

$$27 + W = N, \quad 0.71N = 4 \times 27 + 0.3W$$

Solving the above simultaneous equations results in

$$N = 244\,\text{tons} \quad \text{and} \quad W = 217\,\text{tons}.$$

(2) Ignoring the effect of ^{236}U, we obtain similar material balance equations:

$$N + 25 = 27 + W \quad 0.71N + 0.8 \times 25 = 0.3W + 27 \times 4$$

Solving the above equations results in

$$N = 213\,\text{tons} \quad \text{and} \quad W = 211\,\text{tons}.$$

(3) The amount of natural uranium mined that can be saved by recycling is about 15 %. Although the amount of tailings and waste generated subsequently is expected to be reduced by about the same percentage, uranium recycling is expected to exert only a limited saving effect.

On the other hand, ^{236}U, which was ignored in the above calculation, gets into the product side in the enrichment process together with ^{235}U which has a mass close to that of ^{236}U. ^{236}U absorbs a neutron to become ^{237}U, which then decays into ^{237}Np. In fuel that uses recycled uranium, therefore, a larger amount of ^{237}Np is generated and the toxicity of spent fuel increases significantly.

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An Advanced Course in Nuclear Engineering, DOI 10.1007/978-4-431-55417-2

An approximate calculation is given below to show how ^{237}Np is increased by using recovered uranium. A comparison is made for a PWR with a burn-up of 33 GWd/tHM. The isotopic composition of the recovered uranium is assumed to be the same as that for spent fuel: ^{235}U:^{236}U:^{238}U = 0.92 %:0.41 %:98.67 %. While it is not known how much of the ^{236}U is not separated and gets into fresh fuel together with ^{235}U, let us assume that all ^{236}U enters the fresh fuel. For a ^{235}U isotopic ratio of 4 % in the fresh fuel, the isotopic ratio of ^{236}U is calculated as 4 % × (0.41 %/0.91 %) = 1.8 %.

On the other hand, the amount of recovered uranium in the 27 tons of fresh fuel is 3.4 tons, as derived from the above problem (2) through a simple calculation. For a ^{235}U isotopic ratio of 4 % in the fresh fuel, the isotopic ratio of ^{236}U is calculated as 1.8 % × 3.4 tons/27 tons = 0.23 %. This means 1 tons of fuel contains 2–3 kg of ^{236}U.

In a light water reactor with a burn-up of 33 GWd/tHM, let us assume 868 operating days, a capture cross section of 8.6 b for ^{236}U and an in-core flux of 3.0×10^{14} cm^2/s. The amount of ^{237}Np generated by the neutron capture reaction of ^{236}U is calculated to be about 0.45 kg/ItHM (here, "I" means initial). For the same reactor conditions, the amount of ^{237}Np generated in fresh fuel without ^{236}U is 0.42 kg/ItHM. Accordingly, the use of recovered uranium significantly increases the amount of ^{237}Np generated.

2. (1) The material balance of minor actinides in repetitive transmutation is shown in the table below.

Number of cycles	Minor actinides loaded into a transmutation reactor	Minor actinides discharged from a transmutation reactor	Minor actinides transferred into wastes
1	1	$(1-\alpha)$	$(1-\alpha)\beta$
2	$(1-\alpha)(1-\beta)$	$(1-\alpha)^2(1-\beta)$	$(1-\alpha)^2(1-\beta)\beta$
3	$(1-\alpha)^2(1-\beta)^2$	$(1-\alpha)^3(1-\beta)^2$	$(1-\alpha)^3(1-\beta)^2\beta$
n	$(1-\alpha)^{n-1}(1-\beta)^{n-1}$	$(1-\alpha)^n(1-\beta)^{n-1}$	$(1-\alpha)^n(1-\beta)^{n-1}\beta$

The total amount of minor actinides, x, transferred to wastes is given by

$$\begin{aligned} x &= \sum_{n=1}^{\infty}(1-\alpha)^n(1-\beta)^{n-1}\beta = (1-\alpha)\beta\sum_{n=1}^{\infty}\{(1-\alpha)(1-\beta)\}^{n-1} \\ &= \frac{(1-\alpha)\beta}{1-(1-\alpha)(1-\beta)} \end{aligned}$$

(2) Solving the formula obtained in (1) results in $\beta = \frac{\alpha x}{(1-\alpha)(1-x)}$. From $\alpha = 0.1$ and $x \leq 0.01$, we obtain $\beta \leq 0.001122$. Therefore, the percentage of transfer to the wastes by reprocessing needs to be 0.11 % or less.

Chapter 3

1. As shown in Fig. 3.4, the thermal neutron activation cross section of ^{59}Co is 37.18 b, and the half life of ^{60}Co is 5.27 years.

$$A = N \cdot \phi \cdot \sigma\left(1 - e^{-\lambda t}\right)$$

Because there is 100 ppm (0.01 %) of ^{59}Co in the material, the number of ^{59}Co atoms in a volume of 1 cm^3 is 8.083×10^{19}. The radioactivity concentration of ^{60}Co right after completing a 30 EFPY operation is obtained from

$$A = 8.083 \times 10^{19} \times 1 \times 10^{10} \times 37.18 \times 10^{-24} \times \left(1 - e^{-0.1315 \times 30}\right)$$

as 2.95×10^7 Bq/cm^3. Therefore, the radioactivity concentration of ^{60}Co after a 10-year cooling period is obtained from

$$A' = A \times e^{-\lambda t} = 2.95 \times 10^7 \times e^{-0.1315 \times 10}$$

as 7.91×10^6 Bq/cm^3.

2. Because the piping is cut at 0.75 m intervals including the kerf, a total of 201 circumferential cutting lines occur including both connection ends. With an inner diameter of 297.9 mm, the area of each kerf is $0.2979 \times 3.14159 \times 0.005 = 4.68 \times 10^{-3}$ m^2, and the total area for the 201 lines is 9.41×10^{-1} m^2. The area of the kerf generated by longitudinally halving each pipe section is 150 m × 2 × 0.005 = 1.5 m^2.

Because the ^{60}Co surface contamination density is 1×10^4 Bq/cm^2 and the contamination dispersion ratio is 30 %, the amount of ^{60}Co dispersed in the working space is 7.32×10^7 Bq and the amount of ^{60}Co released from the stack through filters is 7.32×10^3 Bq/y (=8.36×10^{-1} Bq/h). With the relative concentration, therefore, the annual average airborne radioactivity concentration of ^{60}Co at the site boundary becomes 1.84×10^{-10} Bq/cm^3.

The annual internal exposure dose due to inhalation is

$$\begin{aligned}&1.84 \times 10^{-10}\,\mathrm{Bq/cm^3} \times 22.2\,\mathrm{m^3/d} \times 365\,\mathrm{d/y} \times 3.1 \times 10^{-2}\,\mu\mathrm{Sv/Bq}\\ &\quad = 4.62 \times 10^{-8}\,\mu\mathrm{Sv/y}\end{aligned}$$

On the other hand, with a deposition rate of 1.0 cm/s, the surface density of ^{60}Co deposited on the ground surface is 1.84×10^{-6} Bq/m^2. Multiplying by the external exposure conversion factor, we obtain an annual external exposure dose of 4.05×10^{-8} μSv.

3. With a true positive rate of 95 % and a false positive rate of 25 %, we obtain $d' = 2.32$, $b_i = 1350$ cpm × 2 s × 1/60 = 45 counts, $s_i = 2.32 \times 45^{1/2} = 15.6$ counts

and MDCR = 15.6 × 60/2 = 467 cpm. Therefore, with a surveyor efficiency of 0.5, instrument efficiency of 0.24 and surface efficiency of 0.25, we obtain scanMDC as

$$\text{scanMDC} = \frac{467}{\sqrt{0.5} \times 0.24 \times 0.25} = 11,000\,\text{dpm}/100\,\text{cm}^2.$$

Chapter 4

1. In the case of evaluating the ingestion intake exposure through the use of the frying pan, the clearance level is obtained as follows.

The internal exposure dose $D_{ING}(i)$ [Sv/y] due to ingestion intake through the use of the frying pan is given by

$$D_{ING}(i) = C_m(i) \cdot A_f \cdot t_f \cdot R_e \cdot \rho_e \cdot \frac{1 - \exp(-\lambda_i t_i)}{\lambda_i t_i} \cdot D_{CF,ING}(i) \tag{4.13}$$

Here,

$C_m(i)$: radioactivity concentration of nuclide i in the frying pan [Bq/g],
A_f: surface area of the frying pan [m^2],
ρ_e: density of iron [g/m^3],
R_e: iron corrosion rate [m/y],
t_f: annual cooking time involving the use of frying pan,
λ_i: decay constant of nuclide i [y^{-1}],
t_i: exposure time during the exposure scenario [y], and
$D_{CF,ING}(i)$: dose conversion factor for an adult due to ingestion intake of nuclide i [Sv/Bq].

Here, $D_{ING}(i)$ is the reference dose value, 10 μSv/y. The radioactivity concentration $C_m(i)$ of nuclide i in the frying pan is given by

$$C_m(i) = C_{WM}(i) \cdot F_{MC} \cdot T_i(i) \cdot G_M \cdot \exp\left(-\lambda_i t_{pd}\right) \tag{4.14}$$

Here,

$C_{WM}(i)$: radioactivity concentration of nuclide i in the cleared metals [Bq/g],
F_{MC}: weight ratio of the cleared metals in the metals to be recycled,
$T_i(i)$: transfer factor of element i from the recycled metals to the frying pan in the melting process,
G_M: dilution factor for the recycled metals, and
t_{pd}: duration from clearance to recycling [y].

Substituting Eq. (4.14) into Eq. (4.13) and rearranging terms, we obtain the following.

$$C_{WM}(i) = \frac{D_{ING}(i) \cdot \lambda_i t_i}{F_{MC} \cdot T_i(i) \cdot G_M \cdot \exp\left(-\lambda_i t_{pd}\right) \cdot A_f \cdot t_f \cdot R_e \cdot \rho_e \cdot [1 - \exp(-\lambda_i t_i)] \cdot D_{CF,ING}(i)} \tag{4.15}$$

Here, some of the variables in the above equation are given as $A_f = 7.07 \times 10^{-2}$ m^2, $\rho_e = 7.86 \times 10^6$ g/m^3, $R_e = 1.3 \times 10^{-4}$ m/y, $t_i = 1$ y, $F_{MC} = 0.1$, $G_M = 1$ and $t_{pd} = 1$ y. When nuclide i is ^{60}Co, other variables in the above equation are $D_{CF,ING}(i) = 3.49 \times 10^{-9}$ Sv/Bq, $\lambda_i = 0.1315$ y^{-1} and $T_i(i) = 1$ for Co. Because the annual number of cooking hours using the frying pan is 180 h, $t_f = 180 \div (24 \times 365) = 2.1 \times 10^{-2}$. Substituting these variables into Eq. (4.15), we obtain

$$\begin{aligned} C_{WM}(i) = &\left(10 \times 10^{-6} \times (0.1315 \times 1)\right) \\ &\div \left[0.1 \times 1 \times 1 \times \exp(-0.1315 \times 1) \times 7.07 \times 10^{-2} \times 2.1 \times 10^{-2}\right. \\ &\times 1.3 \times 10^{-4} \times 7.86 \times 10^6 \times (1 - \exp(-0.1315 \times 1)) \\ &\left.\times \left(3.49 \times 10^{-9}\right)\right] \\ = &\, 2.3 \times 10^4 \,\mathrm{Bq/g}. \end{aligned}$$

This is the radioactivity concentration of ^{60}Co in the cleared metal corresponding to the reference dose value. The actual clearance level for ^{60}Co, whose determined pathway is external exposure of recycling of wall material and the like (persons to be evaluated: children), is 0.3 Bq/g (See Table 4.1).

2. The external exposure dose due to nuclide i, or $D_{EXT}(i)$ [Sv/y], in the parking lot base course material is given by

$$D_{EXT}(i) = C_m(i) \cdot S \cdot t_f \cdot \frac{1 - \exp(-\lambda_i t_i)}{\lambda_i t_i} \cdot D_{CF,EA}(i). \tag{4.16}$$

Here,

$C_m(i)$: radioactivity concentration of nuclide i in the bitumen [Bq/g],
S: shielding factor for external radiation,
t_f: ratio of hours during which the parking lot is used in a year,
λ_i: decay constant of nuclide i [y^{-1}],
t_i: exposure time during the exposure scenario [y], and
$D_{CF,EA}(i)$: dose conversion factor for external exposure to nuclide i [(Sv/y)/(Bq/g)].

Here, $D_{EXT}(i)$ is the reference dose value, 10 μSv/y. The radioactivity concentration of nuclide i in the bitumen, or $C_m(i)$, is given by

$$C_m(i) = C_{WC}(i) \cdot F_{CC} \cdot G_C \cdot F_{RA} \cdot \exp\left(-\lambda_i t_{pd}\right) \tag{4.17}$$

Here,

$C_{WC}(i)$: radioactivity concentration of nuclide i in the cleared concrete [Bq/g],
F_{RA}: ratio between the bitumen and the recycled coarse aggregate,
t_{pd}: duration from clearance to recycling [y],
F_{CC}: weight ratio of the cleared concrete in the concrete to be recycled, and
G_C: dilution factor for the recycled coarse aggregate

Substituting Eq. (4.17) into Eq. (4.16) and rearranging terms, we obtain the following.

$$C_{WC}(i) = \frac{D_{EXT}(i) \cdot \lambda_i t_i}{F_{CC} \cdot G_C \cdot F_{RA} \cdot \exp\left(-\lambda_i t_{pd}\right) \cdot S \cdot t_f \cdot [1 - \exp(-\lambda_i t_i)] \cdot D_{CF,EA}(i)} \tag{4.18}$$

Here, some of the variables in the above equation are given as $t_i = 1$ y, $G_C = 1$, $F_{CC} = 0.1$, $F_{RA} = 0.25$, $t_{pd} = 1$ y and $S = 1$. When nuclide i is ^{60}Co, other variables in the above equation are $D_{CF,EA}(i) = 1.9 \times 10^{-3}$ (Sv/y)/(Bq/g) and $\lambda_i = 0.1315$ y^{-1}. Because the times of use of the parking lot in a year are 1,000 h, we obtain $t_f = 1{,}000 \div (24 \times 365) = 1.1 \times 10^{-1}$. Substituting these variables into Eq. (4.18), we obtain

$$\begin{aligned} C_{WC}(i) &= \left(10 \times 10^{-6} \times 0.1315 \times 1\right) \div [0.1 \times 1 \times 0.25 \times \exp(-0.1315 \times 1) \\ &\quad \times 1 \times 1.1 \times 10^{-1} \times (1 - \exp(-0.1315 \times 1)) \times 1.9 \times 10^{-3}\big] \\ &= 2.3\,\mathrm{Bq/g} \end{aligned}$$

This is the radioactivity concentration of ^{60}Co in the cleared concrete corresponding to the reference dose value. The actual clearance level for ^{60}Co, whose determined pathway is external exposure of recycling of wall material and the like (persons to be evaluated: children), is 0.3 Bq/g (See Table 4.1).

Chapter 5

1. In this problem we have to consider the interdependence of three processes: treatment (evaporation), conditioning (cementation), and disposal that follows. When carrying out evaporation, because the radioactivity concentration of the liquid wastes to be treated is relatively high, it may be possible that the reference radioactivity concentration for allowing burial disposal is exceeded and an alternative disposal method may have to be selected if concentration is done excessively for volume reduction only. It is therefore necessary to determine the level of concentration by evaporation in consideration of the criteria for burial disposal. We also have to consider the effect of sulfate, which is contained in the liquid wastes, on cement when concentrating the wastes. Moreover, we need to use sulfate-resistant cement rather than ordinary Portland cement in view of long-term stability after disposal.

2. The particulate collection efficiency E of the HEPA filter is given by

$$E = (C_{\mathrm{in}} - C_{\mathrm{out}})/C_{\mathrm{in}}.$$

Because the concentration of radioactive material at the outlet of the first-stage HEPA filter ($C_{\mathrm{out},1}$) is equal to that at the inlet of the second-stage HEPA filter ($C_{\mathrm{in},2}$), that at the outlet of the second-stage HEPA filter ($C_{\mathrm{out},2}$) is given by

$$C_{\mathrm{out},2} = (1 - E)^2 \cdot C_{\mathrm{in},1}.$$

Here, $C_{\mathrm{in},1}$ is the concentration of radioactive material at the inlet of the first-stage HEPA filter. Substituting $C_{\mathrm{in},1} = 0.6$ and $C_{\mathrm{out},2} = 3 \times 10^{-6} \times 1/100$ into the above equation, we obtain the required collection efficiency of the HEPA filter as 99.98 %.

3. Tritium, ^{3}H, which is a volatile radioactive material, is mostly present as vapor in the exhaust generated by incineration. Because it cannot be removed by HEPA or other particulate filters, it needs to be removed by rinsing the off-gas with water. As a result, liquid waste containing tritium is generated as secondary waste. Because tritium cannot be removed by evaporation, filtration, coagulation-sedimentation or ion exchange, the waste, before being discharged, needs to undergo direct cementation or, if the tritium concentration is low, be diluted to a level below the regulation limit for discharge. On the other hand, ^{60}Co can be removed by HEPA or other particulate filters because it is present in the off-gas as particulates. As a result, spent filters are generated as secondary waste. The filters, which are generally incombustible, need to be treated by compacting or melting at high temperatures.

Polyvinyl chloride, whose calorific value is greater than that of cellulosic waste, causes a greater pressure variation in the incinerator as it is combusted. Because the combustion of polyvinyl chloride generates acidic gases, dioxins and other hazardous substances in the incinerator or the environment, preventive measures are required such as combusting polyvinyl chloride as a mixture with cellulosic waste and/or installing equipment for removing hazardous substances.

4. In the compaction treatment, it is generally not necessary to strictly segregate the waste to be treated, and therefore the work load for waste segregation at the pretreatment stage is reduced. There is also an economic advantage that the treatment system is relatively simple and the cost of treatment is relatively low. As a disadvantage, on the other hand, the volume reduction factor of the compaction treatment is lower than that of the melting.

In the melting, segregation is necessary at the pretreatment stage because the type of waste that can be treated depends on the method of heating. Moreover, to reduce the volume of wastes that cannot be treated with the prepared melting system, another volume reduction system needs to be introduced. There is also an economic disadvantage that the melting is generally costly. On the other hand, the density of waste can be reduced to the real density of the waste material. There are also advantages in terms of burial disposal; it is easier to conduct waste characterization including evaluation of radioactivity concentration because the wastes can be homogenized, and it is possible to reduce the leakage rate of radioactive materials.

5. Gaseous wastes may be generated in a nuclear power plant in the following manner. ^{16}N and other radioactive materials generated by the activation of coolant, as well as fission products such as noble gases and iodine released through microdamage in fuel cladding may get into the reactor cooling water. In the case of a BWR plant, these radioactive materials can migrate to the main steam system or leak through pumps, valves or other components. The main radionuclides in the gaseous wastes include ^{131}I, ^{87}Kr, ^{133}Xe, and ^{16}N. The gaseous waste is treated by such systems as charcoal filters and noble gas hold-up systems.

Gaseous wastes generated in a reprocessing plant contain fission products that were produced in spent fuel, and the activation products generated in reactors are not of concern. Also, short-lived nuclides are not of concern because they decay during the cooling period before the start of reprocessing. Of concern in a reprocessing plant are ^{85}Kr, which is a long-lived noble gas nuclide, and ^{129}I. At present, no special treatment is conducted for ^{85}Kr, except for exhaust with ventilation air, which is expected to have a dispersion effect. As for ^{129}I, alkaline cleaning and solidification using a silver adsorbent are being conducted.

6. Vitrification is used as a technique to solidify high-level radioactive liquid wastes for the following reasons.

1. Many of the elements contained in high-level radioactive liquid wastes can be made into a solid solution or dispersed homogeneously. The chemical structure of borosilicate glass for vitrified wastes is a network structure having its main components Si and B bonded through O. In the vitrified wastes, radionuclides in high-level radioactive wastes are homogeneously and stably taken into the network structure.
2. Vitrified waste forms are stable both physically and chemically (thermal stability, resistance to leaching and resistance to radiation). Glass is highly insoluble in water, resistant to alteration over an extended time period, and therefore suitable for confining radioactive materials for a long period.
3. Manufacturing technologies that have been accumulated over many years in the conventional glass-making industry are available for use.

Chapter 6

1. There are three main roles in natural barriers (see reference 10 in Chap. 6): (1) to keep some distance between the wastes and the accessible environment; (2) to maintain an environment suitable for the performance of engineered barriers; and (3) to prevent, as natural barriers, the migration of radionuclides in groundwater.

Item (1) implies that a repository site can be selected by avoiding natural and human factors that would adversely affect the stability of the underground environment. It is therefore required that there are no volcanoes or useable underground resources at the site. As for items (2) and (3), it is necessary to confirm that a chemically reducing condition is maintained and the flow of groundwater is slow. In the "Technical Reliability of Geological Disposal of High-Level Radioactive Waste in Japan" report edited by JNC (1999), doses were evaluated for an alternative scenario in which there were no natural barriers. This is equivalent to ignoring migration paths in natural barriers. Even in this case, it was assumed that engineered barriers were in a chemically reducing environment. This reducing environment greatly affects the solubility of radioactive elements and other parameters.

2. The unit of electric power sold is converted to MWd, which is used in the unit of burnup MWd/MTU. Namely, 9159×10^5 MWh is converted into a value based on MWd, where d represents a day.

Then, the volume of generated vitrified waste per capita [cm^3/person] is given by power sold [MWd/y] × ratio of nuclear power × reciprocal of burnup [1/(MWd/MTU)] × reciprocal of generating efficiency × number of generated vitrified waste forms [forms/MTU] × volume of each vitrified waste form [cm^3/form] × reciprocal of population [1/person] × lifetime [year].

$$\frac{9,159 \times 10^5}{24} \times \frac{1}{3} \times \frac{1}{45,000} \times \frac{100}{33.4} \times 1.25 \times 150 \times 10^3 \times \frac{1}{128,000,000} \times 80 = 99.2$$

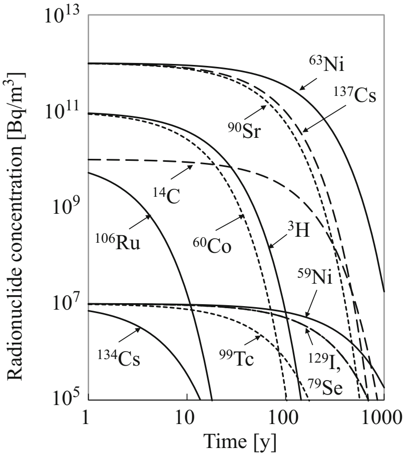
The volume per capita is therefore calculated to be 99.2 cm^3/person; this means about 100 cm^3 (0.1 L) for each person.

Chapter 7

1. Solving Eq. (7.5) with the initial condition of $C_s(0) = C_0$, we obtain the radioactivity concentration of the seepage water in the vault by

$$C_s(t) = C_0 \exp\left[-\left(\lambda + \frac{\nu}{L\theta_s R_s}\right)t\right].$$

Substituting the given conditions into this equation and plotting against time, we obtain the diagram below. The table shows the decay constant of each nuclide and the constant indicating the ratio of the nuclide that flowed out of the vault ($\nu/L\theta_s R_s$). The table explains why ^{129}I and ^{79}Se follow exactly the same curve.



Nuclide	^{3}H	^{14}C	^{60}Co	^{59}Ni	^{63}Ni	^{79}Se
λ	0.0561	1.21×10^{-4}	0.131	9.24×10^{-6}	6.93×10^{-3}	1.07×10^{-5}
$\nu/L\theta_S R_S$	0.0400	0.0133	3.64×10^{-3}	4.00×10^{-3}	4.00×10^{-3}	6.67×10^{-3}

Nuclide	^{90}Sr	^{99}Tc	^{106}Ru	^{129}I	^{134}Cs	^{137}Cs
λ	0.0241	3.25×10^{-6}	0.642	4.41×10^{-8}	0.336	0.0230
$\nu/L\theta_S R_S$	4.44×10^{-3}	0.0267	3.96×10^{-4}	6.67×10^{-3}	3.96×10^{-4}	3.96×10^{-4}

2. The straight line in the figure is given by Eq. (9.57) in Chap. 9 “Solutions to Diffusion Equations and Advection-Dispersion Equations for Radioactive Materials”:

$$\frac{C_{\text{low}}(t)}{C_0} = \frac{AD}{VL}\left(t - \frac{L^2 R}{6D}\right).$$

With the slope of the line $AD/VL = 0.011$ MBq/(m^3·day), the point of intersection with the time axis $LR^2/6D = 2.5$ day, diffusion area $A = 3.14 \times 10^{-4}$ m^2, volume of solution in the cell $V = 1.1 \times 10^{-4}$ m^3 and sample thickness $L = 12$ mm, the diffusion coefficient and retardation factor are calculated to be $D = 5.4 \times 10^{-10}$ m^2/s and $R_s = 12$, respectively. Furthermore, with $R_s = 1 + [(1-\varepsilon)/\varepsilon]\rho K_d$, porosity $\varepsilon = 0.4$ and density $\rho = 1.6 \times 10^3$ kg/m^3, the sorption distribution coefficient is calculated to be $K_d = 0.0046$ m^3/kg.

3. (Omitted)

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