NUCLEAR WASTE MANAGEMENT AND THE NUCLEAR FUEL CYCLE

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Summary

In addition to a rapidly growing demand for more electricity, especially in Asia, concerns over energy resource availability, climate change, air quality, and energy security suggest a larger and more important role for nuclear power in the future. However, it is unlikely the public will accept growth of nuclear power until issues associated with nuclear waste management, reactor safety, economics, and non-proliferation are addressed by both the nuclear industry and government. In this chapter,

the management of nuclear waste from power generation is described in terms the three major fuel cycle options that are under consideration by various countries around the world. The importance of the actinide elements and certain fission products as the long term waste issues is discussed along with both aqueous and non-aqueous separation technologies that can be used to partition these from spent fuel for subsequent transmutation using reactor or accelerator-driven devices in an advanced fuel cycle scenario. The strategy and challenges associated with the deployment of Generation IV Energy systems currently under development are also discussed.

1. Introduction

Radioactive waste, like other wastes, may be composed of materials varying in origin, chemical composition, and physical state. However, what differentiates radioactive waste from other waste forms is that it contains components that are unstable due to radioactive decay. Managing radioactive waste requires different approaches to ensure the protection of both humans and the environment from the radiation.

In general, three options exist for managing radioactive waste: (1) concentrate and contain (concentrate and isolate the wastes in an appropriate environment); (2) dilute and disperse (dilute to regulatory-acceptable levels and then discharge to the environment); and; (3) delay to decay (allow the radioactive constituents to decay to an acceptable or background level). The first two options are common to managing non-radioactive waste but the third is unique to radioactive waste. Eventually all radioactive wastes become benign because they decay to stable elements while non-radioactive, hazardous waste remains hazardous forever or until their chemical speciation is changed

By far the largest source of radioactive waste from the civilian sector results from the generation of power in nuclear reactors. Much smaller quantities of civilian radioactive waste result from use of radionuclides for scientific research as well as from industrial sources such as medical isotope production for diagnostic and therapeutic use and from X-ray and neutron sources. The other significant sources of radioactive waste are due to defense related activities that support the production and manufacture of nuclear weapons. This chapter deals primarily with the management of civilian nuclear waste and focuses on that generated from nuclear power.

2. Classification of Radioactive Wastes

In order to manage nuclear wastes, it is useful to classify or group them into categories based on the properties of the waste, which can be done in a number of ways. For example, radioactive waste can be classified by the level of radioactivity present (high, intermediate, low, or below regulatory concern), by the dominant type of radiation emitted (alpha, beta, gamma, or X-ray) or, by its half-life (a length of time required for the material to decay to half of its original value). Also, radioactive wastes can be classified by their physical characteristics (primarily, solid or liquid, but they can also exist in the gaseous state). A quantitative way to classify radioactive waste is by specific activity or activity concentration; i.e., by the activity per quantity of waste (mass or volume). The heat generated in a sample (that depends on half-life, concentration, and type of radiation) can also be used for classification. Finally, waste

can be classified for security and non-proliferation purposes (e.g., designated as "special nuclear materials"), for worker safety, and for transportation.

These various classifications have advantages and disadvantages depending upon how the information is used. In the United States and many places around the world, radioactive wastes have traditionally been classified on the basis of their characteristics and how the waste was produced via a top down, generator-oriented approach. This has resulted in inconsistencies, overlaps, and omissions which lead to conflicts between the source-defined classifications and the waste acceptance criteria developed from the disposal systems. Such generator-oriented waste classifications do not fully capture the associated hazards and, thus, are insufficient to ensure public health and environmental safety. Whatever qualitative framework is used, the length of time radioactive waste must be isolated from the public is determined primarily by its half-life and energy. The heat generation, concentration, and type of radiation determine the shielding and handling requirements for the disposal of the waste.

Although the classification of radioactive wastes varies from country to country, three groupings are generally accepted internationally.

2.1. High-level Waste (HLW)

HLW generally refers to the radioactive nuclides at high levels from nuclear power generation, (i.e. reprocessing waste streams or unprocessed spent fuel) or from the isolation of fissile radionuclides from irradiated materials associated with nuclear weapons production. When the spent nuclear fuel from reactor operations (civilian or defense) is chemically processed, the radioactive wastes include nuclides from the aqueous phase from the first extraction cycle (and other reprocessing waste streams) as it contains high concentrations of radioactive fission products. As a result, HLW is highly radioactive, generates a significant amount of heat, and contains long-lived radionuclides. Typically these aqueous waste streams are treated by the principle of "concentrate and contain," as the HLW is normally further processed and solidified into either a glass (vitrification) or a ceramic matrix waste form. Spent nuclear fuel not reprocessed is also considered as HLW. Because of the highly radioactive fission products contained within the spent fuel, it must be stored for "cooling" for many years before final disposal by isolation from the environment. This final disposal of HLW is placement within deep geologic formations. Relative to the total volume of waste produced from commercial power generation, HWL constitutes only a small fraction (a few percent). However, the vast majority of the radioactivity (> 95%) resides in the HLW. The only disposal option for this class of waste is burial in a deep geologic repository.

2.2. Intermediate-level Waste (ILW)

ILW contains lower amounts of radioactivity than HLW but still requires use of special shielding to assure worker safety. Reactor components, contaminated materials from reactor decommissioning, sludge from spent fuel cooling and storage areas, and materials used to clean coolant systems such as resins and filters are generally classified as ILW. The most common management option is "delay to decay" for short-lived solid

waste, but for the long-lived waste, the "concentrate and contain" principle (solidification for deep geologic disposal) is required. ILW comprises about 7% of the volume and, roughly, 4% of the radioactivity of all radioactive wastes. The disposal options for this class of waste are burial in a deep geologic repository for the long-lived radionuclides and near-surface burial for the short-lived ones.

2.3. Low-level Waste (LLW)

All civilian and defense-related facilities that use or handle radioactive materials generate some LLW. These include research laboratories, hospitals using radionuclides for diagnostic and therapeutic procedures, as well as nuclear power plants. LLW includes materials that become contaminated by exposure to radiation or by contact with radioactive materials. Items such as paper, rags, tools, protective clothing, filters and other lightly contaminated materials that contain small amounts of short-lived nuclides are usually classified as LLW. By its nature, LLW does not require shielding during normal handling and transportation and both principles of "delay to decay" and "dilute and disperse" can be employed for disposal depending on the exact nature of the waste. Often, it is advantageous to reduce the volume of LLW by compaction or incineration before disposal. Worldwide it constitutes ~90% of the volume but only ~1% of the radioactivity associated with all radioactive waste. However, wastes containing small amounts of long-lived radionuclides can be included under the LLW classification. The disposal options for this class of waste are near-surface burial or no restrictions depending on level of radioactivity.

The International Atomic Energy Agency (IAEA) noted that these classifications, HLW, ILW and LLW, although useful for some purposes, have some important limitations. Specifically the limitations cited are no "clear linkage to the safety aspects in radioactive waste managements, especially disposal," the "current classification system lacks quantitative boundaries between classes," and no "recognition of a class of waste that contains so little radioactive material that it may be exempted from control as a radioactive waste." To overcome these limitations, the IAEA proposed a modified system shown in Figure 1.

In this simple but eloquent scheme, the principal waste classes are exempt waste (explained in the next paragraph), low and intermediate level waste (which may be subdivided into short and long-lived waste and alpha-bearing waste) and high level waste. On the y-axis the radioactivity ranges from very low (simple and conventional disposal) to very high (isolation in a geological repository). As the radioactivity increases so does the heat content, the need for shielding, and need for better and longer isolation from the biosphere. On the x-axis, the decay periods range from very short (seconds) to very long (millions of years) and the associated wastes range in composition from those containing small to significant quantities of long-lived radionuclides.

Exempt waste, also identified as "below regulatory concern" or "*de minimis*", contains very low concentrations of radioactivity. The associated radiological hazards are generally considered to be negligible as they are less than for naturally-occurring radioactivity.

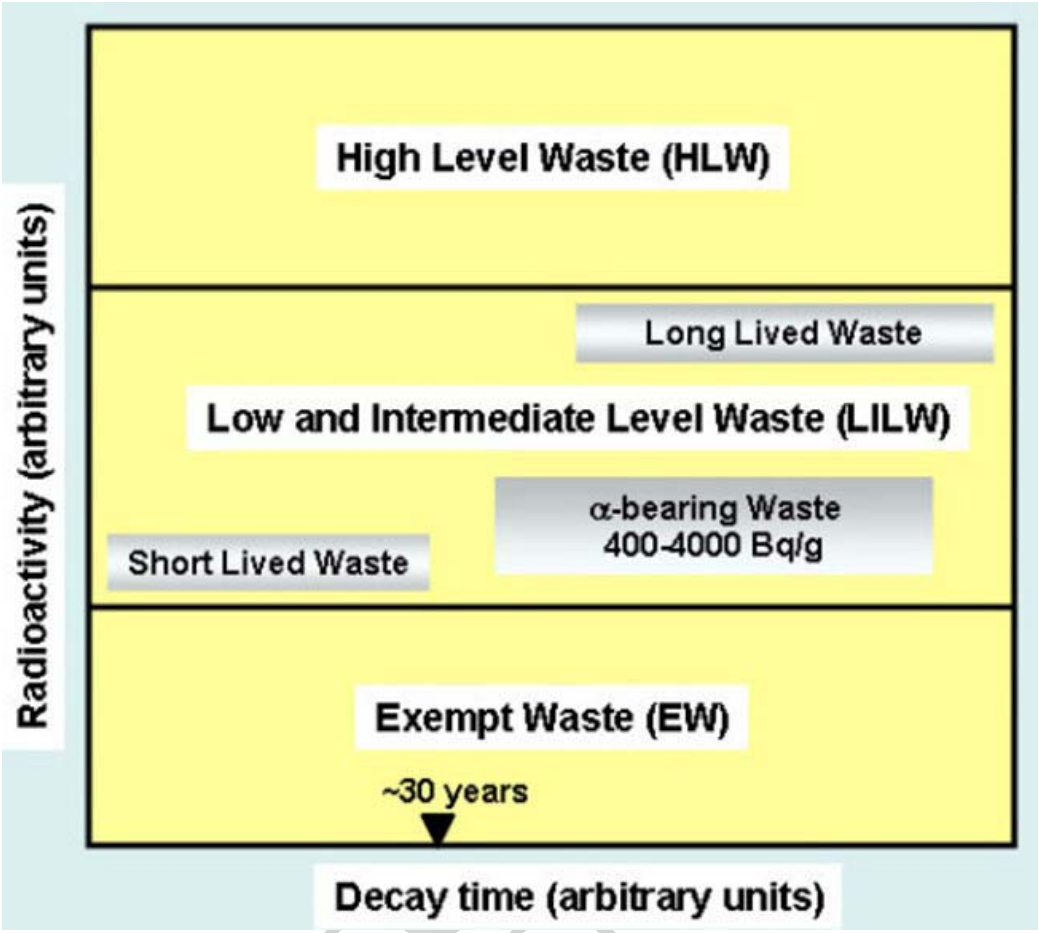

Figure 1: From the IAEA publication: Classification of Radioactive Waste, A Safety Guide, Safety Series No 111-G1.1 (1994).

To date, the classification scheme proposed by the IAEA has not had universal acceptance although this scheme or slight variations of it are in use in a number of countries. For example, most countries recognize and accept exempt waste as the fourth principal classification along with HLW, ILW, and LLW.

In the United States waste classifications are defined by the US Nuclear Regulatory Commission (NRC):

1. High Level Waste (HLW) – (A) The highly radioactive material resulting from the reprocessing of spent nuclear fuel, including liquid waste produced directly in reprocessing and any solid material derived from such liquid waste that contains fission products in sufficient concentrations; and (B) other highly radioactive material for which the NRC, consistent with existing law, requires permanent isolation;
2. Spent Nuclear Fuel (SNF) – Fuel that has been withdrawn from a nuclear reactor following irradiation, the constituent elements of which have not been separated by reprocessing; the NRC requires SNF be regulated as HLW;
3. Transuranic (TRU) Waste – Waste contaminated with alpha-emitting transuranic elements ($Z > 92$) with half-lives greater than 20 years and in concentrations

higher than 100 nanocuries/gram (3700 Bq g^{-1});
4. Uranium mining and mill tailings – Tailings or wastes produced by the extraction or concentration of uranium or thorium from any ore processed primarily for its source material content (also called by-product material);
5. Low-Level Waste (LLW) – Radioactive material that is not high-level radioactive waste, spent nuclear fuel, nor by-product material; low level waste is further subdivided as shown in Table 1;
6. Naturally Occurring and Accelerator produced Radioactive Materials (NORM/NARM).

LLW Waste Class	Definition
Class A	Low levels of radiation and heat, no shielding required to protect workers or public, rule of thumb states that it should decay to acceptable levels within 100 years
Class B	Higher concentrations of radioactivity than Class A and requires greater isolation and packaging (and shielding for operations) than Class A waste.
Class C	Requires isolation from the biosphere for 500 years. Must be buried at least 5m below the surface and must have an engineered barrier (container and grouting)
Greater than Class C	This is the LLW that does not qualify for near-surface burial. This includes commercial transuranics (TRUs) that have half-lives > 5 years and activity > 100 nCi/g.

Table 1: Subclasses of low-level waste according to the US Nuclear Regulatory Commission (US NRC) - U.S. Code of Federal Regulations, Title 10, Part 61 (10 C.F.R. 61).

3. Who is Responsible for Radioactive Wastes?

Internationally it is accepted that each country is ethically and legally responsible for the radioactive wastes generated within its borders. Specifics related to what constitutes radioactive waste, the parties responsible, as well as the bodies charged with regulating its use and ultimate disposal, are issues defined by governmental processes.

3.1. Pertinent Legislation in the US Regarding Radioactive Wastes: An Example

In the United States, legislation has resulted in laws that govern the production, use, and means of disposal of radioactive materials. In 1946 the McMahon Energy Act created the Atomic Energy Commission (AEC) with a charter to operate the Manhattan Project facilities. The Atomic Energy Act of 1954, as amended, defined responsibilities related to production, use, ownership, liability, and disposal. This legislation also made possible the creation of the civilian nuclear power program within the US. It was not until about 1970 that the AEC defined high level waste. HLW was defined as aqueous

waste resulting from solvent extraction cycles associated with reprocessing spent reactor fuel. Four years later, Congress enacted the Energy Reorganization Act that split the AEC into two organizations, the Energy Resource and Development Administration (ERDA) and the Nuclear Regulatory Commission (NRC). ERDA, now the Department of Energy (DOE), was given the task of developing and promoting nuclear power while the NRC was tasked with licensing and regulating the emerging nuclear power industry. In 1980, the Low-Level Radioactive Waste Policy Act and an amendment in 1985, transferred responsibility for disposal of non-defense related low level waste from the federal government to the states but kept commercial transuranic (TRU) waste and certain low-level wastes containing long lived radionuclides within the responsibility of the federal government. This legislation also mandated that the Environmental Protection Agency (EPA) establish radiation protection from LLW. Disposal of HLW and spent fuel was the focus of the Nuclear Waste Policy Act (NWPA) in 1982 which called for the construction of two permanent repositories on the west and east coasts. The NWPA was amended in 1987 to name Yucca Mountain as the only site that would be developed as a repository.

The disposal of HLW remains a major public policy issue in the US as well as abroad. Public concerns and political pressures have led to the need to demonstrate the feasibility and safety of HLW disposal and, in some countries, to implement laws that require operational HLW disposal capability in the next 10–50 years. Many countries, including the United States, have programs dedicated to the disposal of HLW. However, all HLW produced to date is being stored and no permanent disposal system has been approved.

4. Splitting the Atom for Energy

Nuclear power produces energy by fissioning nuclides, most notably ^{235}U or ^{239}Pu. When these nuclides fission, the resulting fission fragments and neutrons have a combined mass less than the parent nuclide. During the fission process, the difference in mass is converted to energy and results in the release of about 200 MeV of energy (or 3.2×10^{-11} joules) for every atom that fissions. The partitioning of the energy resulting from the fission of ^{235}U is shown in Table 2.

	Description	**Approx. Energy (MeV)**
Prompt Energy	Kinetic energy of fission fragments	164.5
	Kinetic energy of prompt neutrons (2.5 neutrons/fission event with a average energy of ~2 MeV)	5.0
	prompt γ-rays	7.0
	Subtotal	176.50
Delayed Energy	Kinetic energy of β-particles emitted in decay of fission products	6.5
	Neutrinos	10.5

	γ-rays	6.5
	Subtotal	23.5
Total	Energy/fission of an atom of U-235 in MeV	200.0

Table 2: Partitioning of energy in the fission of ^{235}U

A great advantage of nuclear power is its ability to extract a tremendous amount of energy from a small volume of fuel. In comparison to fission, the burning of fossil fuels involves only the breaking of chemical bonds, and as a result, only 4 eV or 6.5×10^{-19} joules/molecule of CO_2 are released in the combustion of a carbon atom. Another advantage is related to the inherent characteristics of the fuel itself. Aside from its potential use in nuclear weapons, uranium ore has little use other than for the production of electricity through nuclear power. In contrast, fossil fuels can potentially be used for a wide variety of applications from the production of pharmaceuticals to various important synthetic materials production and to transportation. A third inherent advantage of nuclear power is that under normal operations, energy is produced with minimal environmental impact in terms of air and water pollution compared with burning of fossil fuel, particularly coal. Fossil fuels release large quantities of CO_2 and sulfur and nitrogen oxides (acid rain) to the atmosphere and substantial amounts of fly ash as solid waste. International concern is increasing over the "Greenhouse Gas" threat from fossil fuel. Energy created by splitting the atom does not produce greenhouse gases.

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Biographical Sketches

Patricia Ann Baisden received her BS degree (with honors) in 1971 and her Ph.D. in chemistry in 1975 under the direction of Prof. Gregory R. Choppin at the Florida State University. After completing her graduate degree, Dr. Baisden accepted a postdoctoral appointment with Nobel Laureate, Glenn T. Seaborg at the Lawrence Berkeley Laboratory. Since 1978, she has worked for the Lawrence Livermore National Laboratory in California holding a number of technical and management positions including Deputy Director of the Glenn T. Seaborg Institute for Transactinium Science and Division Leader, Materials Program Leader, Chief Scientist, and Deputy Associate Director in the Chemistry & Materials Science Directorate. Her expertise is in nuclear and radiochemistry; actinide chemistry, and lasers and optical materials and over her career she has authored or co-authored over 50 journal papers. Dr. Baisden has also held a number of professional positions including serving as Chairman of the American Chemical Society's Division of the Nuclear Chemistry and Technology, as a member of the Department of Energy (DOE)/National Science Foundation (NSF) Nuclear Science Advisory Committee, and as an editor of ***Radiochimica Acta***, the International Journal for the Chemical Aspects of Nuclear Science and Technology. Additionally, she has been appointed to a number of National Academy of Sciences/National Resource Council study committees including the Committee on Environmental. Technologies for Decontamination and Decommissioning, Committee on Electrometallurgical Techniques for DOE Spent Fuel and Excess Weapons Plutonium Disposition; Committee on the Assessment of the National Institutes of Standards and Technology (NIST) Physics Laboratory, and the Chemical Sciences Roundtable. Dr. Baisden is also credited with starting in 1983 the "Summer School in Nuclear Chemistry," an Undergraduate Fellowship Program for DOE. This program, which will be held

for its 25th consecutive year in 2007, has to date, introduced over 550 undergraduate students to the field of nuclear and radiochemistry. Currently Dr. Baisden holds the position of Assistant Associate Director in the National Ignition Facility (NIF) Programs Directorate at LLNL and is on special assignment to the Office of Inertial Confinement Fusion with the DOE/National Nuclear Security Administration in Washington, DC.

Gregory R. Choppin received a Maxima Cum Laude Bachelor of Science degree for Chemistry from Loyola University, New Orleans, Louisiana, in 1949 and a Ph.D. from the University of Texas, Austin, in1953. He then began postdoctoral research with Glenn T. Seaborg in the new element research group at the Lawrence Radiation Laboratory, University of California-Berkeley. In 1955, he accepted a permanent position in the Lawrence Laboratory and was one of the four co-discovers of element 101, Mendelevium. These experiments were the first on single atoms as only one atom of Mendelevium was made per hour of irradiation in the cyclotron. In 1956, he moved to Florida State University as an Assistant Professor of Chemistry. At FSU, he initiated a research program in f-element chemistry, studying both lanthanide (4f) and actinide (5f) behavior. He spent sabbatical years at the Belgian Nuclear Center (Mol) in 1962-63 and at the German Nuclear Center (Karlsruhe) in 1979-80. He has spent shorter periods for research in nuclear laboratories in France, Portugal, Sweden, and Japan. His research has resulted in over 400 journal publications and 8 books. A major focus of his research has been the thermodynamics and kinetics of f-element complexation reactions. Since 1975, he has focused attention increasingly on the environmental behavior of the actinide elements. Recognition of Professor Choppin's research is reflected in a number of awards among which are American Chemical Society Award in Nuclear Chemistry; Presidential Citation Award, American Nuclear Society; Chemical Pioneer Award, American Institute of Chemistry; Award in Chemical Education, Manufacturing Chemists Association; Award in Rare Earth Chemistry, Rare Earth Society, Japan; Becquerel Medal in Radiochemistry, British Royal Society of Chemistry; Honorary Doctor of Science, Loyola University, New Orleans; Honorary Doctor of Technology, Chalmers University, Sweden; Alexander von Humboldt Prize, Germany; Hevesy Medal for Nuclear Research, Hungary. In 1969, he was given the honor of becoming an R.O. Lawton Distinguished Professor at Florida State University. In 2001, Professor Choppin became a Professor Emeritus.