CHAPTER 5 – RADIOACTIVE WASTE MANAGEMENT

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5.1. INTRODUCTION

The ore pitchblende was discovered in the 1750's near Joachimstal in what is now the Czech Republic. Used as a colorant in glazes, uranium was identified in 1789 as the active ingredient by chemist Martin Klaproth. In 1896, French physicist Henri Becquerel studied uranium minerals as part of his investigations into the phenomenon of fluorescence. He discovered a strange energy emanating from the material which he dubbed "rayons uranique." Unable to explain the origins of this energy, he set the problem aside.

About two years later, a young Polish graduate student was looking for a project for her dissertation. Marie Sklodowska Curie, working with her husband Pierre, picked up on Becquerel's work and, in the course of seeking out more information on uranium, discovered two new elements (polonium and radium) which exhibited the same phenomenon, but were even more powerful. The Curies recognized the energy, which they now called "radioactivity," as something very new, requiring a new interpretation, new science. This discovery led to what some view as the "golden age of nuclear science" (1895-1945) when countries throughout Europe devoted large resources to understand the properties and potential of this material.

By World War II, the potential to harness this energy for a destructive device had been recognized and by 1939, Otto Hahn and Fritz Strassman showed that fission not only released a lot of energy but that it also released additional neutrons which could cause fission in other uranium nuclei leading to a self-sustaining chain reaction and an enormous release of energy. This suggestion was soon confirmed experimentally by other scientists and the race to develop an atomic bomb was on. The rest of the development history which lead to the bombing of Hiroshima and Nagasaki in 1945 is well chronicled. After World War II, development of more powerful weapons systems by the United States and the Soviet Union continued to advance nuclear science. It was this defense application that formed the basis for the commercial nuclear power industry.

The Dawn of the Commercial Nuclear Power Industry

Both the Soviet Union and the West realized that the tremendous heat produced in the process could be tapped either for direct use or for generating electricity. It was also clear that this new form of energy would allow development of compact long-lasting power sources which could have various applications. The first nuclear reactor to produce electricity was the Experimental Breeder Reactor (EBR-1) in Idaho, USA December 1951. In 1953 President Eisenhower proposed the "Atoms for Peace" program,

which reoriented significant research effort towards electricity generation and set the course for civil nuclear energy development in the United States. The main US effort was under Admiral Hyman Rickover, which developed the Pressurized Water Reactor (PWR) for naval (particularly submarine) use. The PWR used enriched uranium oxide fuel and was moderated and cooled by ordinary (light) water. The Mark 1 prototype naval reactor started up in March 1953 in Idaho, and led to the US Atomic Energy Commission building the 60 MWe Shippingport demonstration reactor in Pennsylvania, which started up in 1957 and operated until 1982. The Shippingport reactor spurred the commercial nuclear power industry in the USA.

Similar development occurred across the globe and today there are over 400 reactors of varying configurations operating throughout the world with a total installed capacity of over 370 GWe or about 15% of the world's electricity (vs. coal-40%, oil-10%, natural gas-15%, and hydro & other-19%) [1]. Today, the nuclear industry is at the eye of a "perfect storm." Fuel oil and natural gas prices near record highs, worldwide energy demands increasing at an alarming rate, and increased concerns about greenhouse gas (GHG) emissions have caused many to look negatively at long-term use of fossil fuels. This convergence of factors has led to a growing interest in revitalization of the nuclear power industry within the United States and across the globe. The International Atomic Energy Agency (IAEA) revised upwards its projections for 2030 [2]. Its low projection shows an increase from 372 GWe today to 511 GWe in 2030, the high one gives 807 GWe then, in line with higher forecast growth in power generation.

Radioactive Waste Generation

As with any industrial process, commercial nuclear power results in the generation of process wastes. To facilitate communication and information exchange regarding treatment and handling of radioactive wastes, the IAEA instituted a revised waste classification system in 1994 that takes into account both qualitative and quantitative criteria [3]. As defined below, the IAEA developed a system to classify these wastes in three principal classes including Exempt Waste, Low- and Intermediate-Level Waste (LILW), and High-Level Waste (HLW).

> ***Exempt Waste:*** (EW) contains such a low concentration of radionuclides that it can be excluded from nuclear regulatory control because radiological hazards are considered negligible.
>
> ***Low and Intermediate Level Waste:*** (LILW) contains enough radioactive material that it requires actions to ensure the protection of workers and the public for short or extended periods of time. This class includes a range of materials from just above exempt levels to those with sufficiently high levels of radioactivity to require use of shielding containers and in some cases periods for cooling off. LILW may be subdivided into categories according to the half-lives of the radionuclides it contains, with "short-lived" being less than 30 years and "long-lived" greater than 30 years.

> ***High Level Waste:*** (HLW) contains sufficiently high levels of radioactive materials that a high degree of isolation from the biosphere, normally in a geologic repository, is required for long periods of time. Such wastes normally require both special shielding and cooling periods.

Substantial amounts of radioactive waste are generated through civilian applications of radionuclides in medicine, research and industry. A typical 1000 MWe nuclear power station produces approximately 300 m^3 of low- and intermediate-level waste (LILW) per year and some 30 tons of high-level solid packed waste per year. By comparison a 1000 MWe coal plant produces some 300,000 tons of ash alone per year containing radioactive material and heavy metals which end up in landfill sites and in the atmosphere [4]. Worldwide, nuclear power generation facilities produce about 200,000 m^3 of LILW and 10,000 m^3 of HLW (including spent fuel designated as waste) each year worldwide.

5.2. NUCLEAR WASTE TREATMENT & PROCESSING

Research studies into the management of radioactive waste began in the 1930s. As the commercial nuclear industry evolved through the 1960s and 1970s, additional emphasis was placed on developing long-term solutions for radioactive wastes. Exempt and LILW from commercial nuclear power facilities are handled much like ordinary municipal wastes, although most LILW is disposed of in stable near-surface disposal sites, or as is the case with transuranic waste (TRU) from the United States defense program in stable salt-based repositories (such as the Waste Isolation Pilot Plant, WIPP in New Mexico, USA).

In 2007, in response to growing concerns about management of LILW in the United States, the US Government Accountability Office (GAO) performed a comprehensive review of worldwide practices associated with LILW handling [5]. This report provides a comprehensive analysis of management approaches for LILW, including soil, debris, rubble, process materials, and clothing that have been exposed to radioactivity or contaminated with radioactive material. The report also looked at disposition of excess sealed radiological sources that are no longer useful for industrial or medical applications. The GAO found that most countries maintain waste inventory databases that include information on waste generators (nuclear utilities, hospitals, universities, and research laboratories), waste types, storage locations, and present and future waste generation predictions and disposal capacity needs. The report also found that disposal practices vary according to the hazard presented by the LILW in question. As discussed above, lower-activity LILW is handled much like municipal waste and is typically disposed in near-surface burial sites that are monitored over time. Depending on the level of activity being treated, most of this disposal is handled as Exempt Waste per the IAEA definitions (see above) and no review is required from the nuclear regulatory authority. For higher-activity LILW, most countries have centralized storage and disposal options that are licensed by the appropriate nuclear regulatory authority. Funding for operations of these facilities is either provided by the central government or by collecting disposal fees at the time of disposal. For sealed sources used in industrial and medical applications, the disposal fee is often collected at the time of purchase.

The discussion in the remainder of this chapter will focus on handling of spent nuclear fuel and HLW. A typical 1000 MWe nuclear reactor generates about 25 tons of used fuel each year. In the United States and Canada, this used fuel is regarded as waste and is slated for direct disposal. In most of Europe and Japan, the used fuel is reprocessed to recover unused uranium and to efficiently manage TRU and fission products. In either case, the used fuel is first stored for several years under water in cooling basins at the reactor site (see Figure 1). The water covering the fuel assemblies provides radiation protection, while removing the heat generated during radioactive decay.

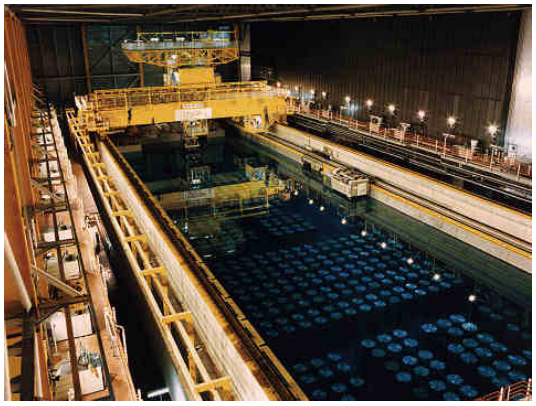

Figure 1. Typical cooling basin for used nuclear fuel.

If the used fuel is reprocessed, the nuclear fuel assemblies are disassembled and chopped into small pieces in a highly-secure, remote processing environment. The fuel core is typically dissolved in nitric acid and separated chemically into uranium, plutonium and high-level waste solutions. About 97% of the used fuel can be recycled leaving only 3% as high-level waste. The resulting hulls from the fuel assemblies are treated as LILW.

For a typical l000 MWe nuclear reactor, about 230 kilograms of plutonium (1% of the spent fuel) is separated in reprocessing annually. The separated Pu can be used in fresh mixed oxide (MOX) fuel or stored for later handling and disposal. MOX fuel fabrication has been ongoing in Europe, with some 25 years of operating experience. A similar plant is scheduled to start-up in Japan in 2012. A MOX fuel plant is also under construction in the United States (at the Savannah River Site in Aiken, SC) for disposition of excess Pu from the United States nuclear weapons program.

Due to the presence of long-lived radionuclides, the separated high-level wastes (about 3% of the typical reactor's used fuel) needs to be isolated from the environment. Research into the processing and disposal of HLW has been active for many years. Isolation of the HLW has typically been in large underground storage tanks. In the United States, about 75 million gallons have been stored in 177 tanks at Hanford, 53 tanks at Savannah River, and two tanks at the West Valley Demonstration Project. The HLW is in the form of a liquid or slurry and needs to be solidified into a waste form before transport to a final storage/disposal facility.

Development of appropriate waste forms began in the United States in the late 1940s at the Brookhaven National Laboratory [6]. The materials studied were based on montmorillonite clay and phosphate glass. France began making borosilicate glass in 1963 [7]. Over the years, a wide variety of materials have been developed and studied for the ultimate isolation and disposal of HLW. The list includes:

- Borosilicate glass
- SYNROC
- Porous glass matrix
- Tailored ceramic
- Pyrolytic carbon and SiC-coated particles
- FUETAP (and other) concretes
- Glass marbles in a lead matrix
- Plasma spray coatings
- Phosphate glass
- Titanate ceramic
- Various calcines

In the late 1970s, the United States Department of Energy (DOE) formed an alternative waste form peer review panel consisting of prominent, independent engineers and scientists with expertise in materials science, ceramics, glass, metallurgy, and geology [8]. Using a rating process to evaluate the relative merit of all the various proposed waste forms, the panel selected borosilicate glass as the reference waste form.

Borosilicate glass is now the material of choice for incorporating and immobilizing the potentially hazardous radionuclides in HLW. Factors that contribute to the suitability of glass waste forms fall into two main categories. First, glass waste forms possess good product durability. Various glass systems are able to incorporate a variety of waste compositions into durable waste forms. These forms have demonstrated good chemical and mechanical performance as well as good radiation and thermal stability. Second, waste-glass forms possess good processing characteristics. The technology for making waste-glass forms is both well developed and well demonstrated. Waste-glass forms ranging in size from bench- and laboratory-scale products to multi-ton canisters have been successfully produced by using ceramic melters as well as in-can melting techniques. The vitrification process provides a substantial volume reduction; a piece the size of a hockey puck would contain the total HLW arising from nuclear electricity generation for one person throughout a normal lifetime (see Figure 2, [9]). Vitrification has been used for nuclear waste immobilization for more than 40 years across Europe, Japan, and the USA.

Figure 2. Approximate size of borosilicate HLW glass produced from nuclear electricity generation over a human lifespan.

In a typical processing facility, the HLW solution is combined with borosilicate glass forming materials and sent to a Joule-heated ceramic melter. Operating at ~1200 °C, the melter turns the waste slurry into molten glass which exits the melter into large stainless steel canisters. An international reference stainless steel canister holds approximately 400 kg of waste glass. After filling, the canisters are decontaminated and sealed by welding a steel plug into the neck of the canister. In most cases, the canisters are temporarily stored at the vitrification facility prior to eventual transport to a geological repository.

Cement and cement-based materials are used to contain by-products from reprocessing operations that are categorized as LILW. Common advantages of cement stabilization include; continuous or batch processing at ambient temperatures, low-cost raw materials, suitability for large or small volumes of many different waste types, and ability to use modular equipment.

Waste stabilization/solidification is most commonly accomplished by mixing aqueous-based wastes with hydraulic or pozzolanic materials such as Portland cements, calcium aluminate cements, calcium sulfo-aluminate (CSA) cements, magnesium (aluminum) phosphate cements, kiln dusts, fly ashes and reactive slags. These materials react with water to form insoluble binders. Composite cement systems using several of these phases are commonly used by the nuclear industry. The hydrated binder phases encapsulate solid particles in the waste, co-precipitate selected contaminant species, and adsorb excess water and soluble contaminants. In addition, the aqueous chemistry of the cement-waste mixture can be adjusted so that the soluble contaminants are precipitated from solution simultaneously with the formation of the matrix phases. Mixtures of the cementitious ingredients plus other additives such as sodium silicate (hardening agent), set accelerators and retarders are commonly used. As a result, a monolithic waste form can be produced at ambient temperatures. The waste forms can also be designed to have a wide range of properties. Compressive strengths typically range from 50-3000 psi. Viscosity and set time can also be adjusted to meet mixing and placement requirements dictated by the production process. Composite cements are typically associated with a highly alkali environment that is not suitable for all wastes; Al metal for example will corrode in such an environment. As a result, a toolbox of cement systems is being developed including geopolymers, CSA cements, and alkali activated systems with at least one suitable for all waste types [10].

Hydrated waste forms are typically used for stabilizing aqueous wastes, such as, condensed off gas wastes, electroplating sludges, salt solutions, incinerator ash, electrostatic precipitator and bag house wastes, and process residues, such as, metal chloride and hydroxide bottoms from ore refining processes. Cementitious materials are also used in a variety of environmental remediation actions to stabilize seepage basin sludges, contaminated soils, and waste disposal sites. In addition, cement-based materials are also used for underground waste tank and pipeline closures. The standard requirement for this application is subsidence prevention. Portland cement based grouts or pumpable, self-leveling, self-compacting backfills containing Portland cement are typically used for tank stabilization. Special grout or backfill formulations are also being designed to stabilize residual contaminants which may not have been removed from these tanks.

5.3. GEOLOGIC DISPOSAL

As discussed above, the HLW fraction of the radioactive waste will require geological isolation for extended periods of time (up to 1,000,000 years). When dealing with such extended time-periods, the fate of the stabilized waste is often questioned. In fact, there is a natural example that suggests that final disposal of HLW underground is safe. Over two billion years ago at Oklo in Gabon, West Africa, chain reactions started spontaneously in concentrated deposits of uranium ore. Scientists estimate that these natural nuclear reactors continued operating for hundreds of thousands of years, forming plutonium and the other by-products created today in a nuclear power reactor. This same area remained highly saturated following the end of the nuclear reaction. Evidence shows that the materials that would be classified as HLW using the current IAEA guidelines remained where they were formed and eventually decayed into non-radioactive elements; they were not mobile in the environment. It is this natural analog that provides the basis for geological isolation of HLW and/or spent nuclear fuel.

Initial discussions regarding a permanent solution to the disposal of high-level radioactive waste in the United States culminated in a report by the National Academy of Sciences – National Research Council in 1957 [6]. The primary recommendation was that "disposal in salt is the most promising method for the near future." Other report recommendations included stabilization of the waste in a "slag or ceramic material forming a relatively insoluble product," the potential of disposing of the waste in a deep repository, the separation of Cs^{137} and Sr^{90}, and consideration of the transportation costs.

A generation later, a similar report [11] expanded on the original as a result of the research completed in the interim. This report concluded that the technology for geologic disposal "is predicted to be more than adequate for isolating radioactive wastes." Other recommendations examined the criterion for system performance, repository design and construction, waste package design, prediction of the performance of the system, and expanded the list of potential host-rock candidates to basalt, granite, salt, and tuff.

Following this report, the United States enacted a policy to dispose of used nuclear fuel and HLW from defense applications at Yucca Mountain, Nevada. The US maintained this

policy for over 20 years and submitted a License Application for the Yucca Mountain Repository to the US Nuclear Regulatory Commission (NRC) in 2008. Recently, however, there has been a Presidential decision to cancel the Yucca Mountain project and reevaluate disposal alternatives in the United States. A recent paper [12] compared the repository programs of the United States and Sweden. In addition to examining the technological aspects of geologic disposal, this report discusses the regulatory and bureaucratic aspects of the two programs and how those issues affect the success or failure of overall nuclear waste management systems.

The experience in the United States proves that an essential aspect of the waste isolation strategy is that long-term safety of geologic disposal must be convincingly presented, and accepted, long before a repository can be opened [13]. As discussed above, this requires safety assessments that consider timescales far beyond the normal horizon of societal thinking. For example, it must be acknowledged that the most robust and passively safe system that can be devised by current generations may ultimately be compromised by the actions of a future society, through inadvertent intrusion [13]. These probabilities must be taken into account in assessing the performance of the repository throughout its operational lifetime. Finally, the scientific analysis and decision-making process must involve stakeholders (i.e., regulators, political leaders, public interest groups and other non-government entities) at local-, regional-, and national-levels.

5.4. CONCLUSIONS

Expansion of nuclear energy worldwide is necessary to meet increased power demands and greenhouse gas (GHG) reduction targets. Virtually any international energy forecast predicts large growth in nuclear power generation over the next 30-50 years. The expansion of commercial nuclear power production will result in increased radioactive waste generation. Managing this waste effectively is a critical component of a world-wide "nuclear renaissance" and will continue to be a primary consideration for future nuclear fuel cycles. Past practice has proven the wastes generated from nuclear power generation can be effectively managed using technologies that are well demonstrated at industrial scales, with decades of safe operating practices. As the worldwide community continues to investigate advanced nuclear fuel cycles, radioactive waste management considerations are being given increased emphasis and are, in fact, being considered at the advent of proposed fuel cycles. This emphasis is necessary to ensure that future nuclear fuel cycles remain economically and *environmentally* competitive (as compared to other forms of energy production) and do not produce legacy waste management issues for future generations.

REFERENCES

1. World Nuclear Association, *World Energy Needs and Nuclear Power,* see http://www.world-nuclear.org/info/inf16.html *(*2009).
2. International Atomic Energy Agency, *Energy, Electricity and Nuclear Power Estimates for the Period to 2030*, IAEA Reference Data Series No. 1, 2009 Edition, Vienna, Austria (2009).

3. International Atomic Energy Agency, *Classification of Radioactive Waste: A Safety Guides,* SAFETY SERIES No. 111-G-1.1, Vienna, Austria (1994).
4. Alex Gabbard, *Coal Production: Nuclear Resource or Danger,* ORNL Review, Volume 26, 3/4, Oak Ridge National Laboratory, Oak Ridge, Tennessee USA (1993).
5. United States Government Accountability Office, *Low-Level Radioactive Waste Management: Approaches Used by Foreign Countries May Provide Useful Lessons for Managing U.S. Radioactive Waste,* GAO-07-221, US Government Accountability Office, Washington, DC, USA (2007).
6. United States National Academy of Sciences – National Research Council, *The Disposal of Radioactive Waste on Land,* Report of the NRC Committee on Waste Disposal of the Division of Earth Sciences, September (1957).
7. Yves Sousselier, *Use of Glasses and Ceramics in the French Waste Management Program,* Page 9 in *Ceramics in Nuclear Waste Management*, CONF-7990420, T. D. Chikalla and J. E. Mendel, editors, (1979).
8. L. L. Hench, *The 70's: From Selection of Alternative Waste Forms to Evaluation Storage System Variables,* Page 129 in *Environmental Issues and Waste Management Technologies in the Ceramic and Nuclear Industries*, Ceramic Transactions Volume 61, V. Jain and R. Palmer, editors (1995).
9. World Nuclear Association, *Waste Management,* see http://www.world-nuclear.org/education/wast.htm *(*2007).
10. N.B. Milestone, *Reactions in Cement Encapsulated Nuclear Wastes: Need for a Toolbox of Different Cement Types,* Adv. Appl. Ceramics **105** [1] 13-20 (2006).
11. T.H Pigford, "The National Research Council Study of the Isolation System for Geologic Disposal of Radioactive Wastes," in The Technology of High-Level Nuclear Waste Disposal, Peter L. Hofmann, editor (1987).
12. L.G. Eriksson, "Spent Fuel Disposal – Success vs. Failure," Radwaste Solutions, January/February, Page 22 (2010).
13. Organization for Economic Cooperation & Development, Nuclear Energy Association, *The Environmental and Ethical Basis of Geological Disposal of Long-Lived Radioactive Wastes: The Geological Disposal Strategy for Radioactive Wastes,* OECD Publications, 2, rue André-Pascal, 75775 Paris Cedex 16, France (1995).