Geological Disposal of Nuclear Waste: a Primer

Rodney C. Ewing[1], Robert A. Whittleston[2], and Bruce W.D. Yardley[3]

ABSTRACT

The back-end of the nuclear fuel cycle has become the Achilles Heel of nuclear power. After more than 50 years of effort, there are, at present, no operating nuclear waste repositories for the spent nuclear fuel from commercial nuclear power plants or for the high-level waste from the reprocessing of spent fuel. This issue describes the status of geological disposal in salt, crystalline rock, clay, and tuff, as presently developed in five countries.

KEYWORDS: nuclear waste, nuclear power, spent nuclear fuel, vitrified high-level nuclear waste, geological disposal, multiple barriers

For the past 50 years there has been an international effort to develop deep-mined geological repositories for the disposal of nuclear waste. This issue of *Elements* presents the reader with selected examples of the approaches that have been developed for different rock types—clay, salt, crystalline rock, and volcanic tuff. For each rock type, the scientific and engineering strategies are different and driven by the properties of the different geological formations, the types of waste to be disposed of, and the regulatory requirements of each country.

The earliest discussions of solutions for the disposal of nuclear waste date from the mid-1950s, and the first US National Academy of Sciences (NAS) Committee on Waste Disposal report (NAS–NRC 1957). The basic principles were reaffirmed 20 years later (NAS–NRC 1978). Because radioactive waste retains potentially harmful

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1 Department of Geological Sciences
Stanford University
Stanford, CA 94305, USA
E-mail: rewing1@stanford.edu
2 European Nuclear Research Centre
Hitachi Europe Ltd
London, UK
E-mail: rob.whittleston@gmail.com
3 Radioactive Waste Management
Building 587, Curie Avenue
Harwell Oxford
Didcot, UK
E-mail: bruce.yardley@nda.gov.uk

levels of radioactivity for hundreds of thousands of years, geological disposal has been preferred from the outset. The first thoughtful scientific review by Earth scientists of what would be required was by John Bredehoeft and colleagues in 1978. This prescient paper recognized the importance of different waste types, the challenges of site characterization, the perturbations that the emplaced waste would impose on geological formations, and the time frames over which geological isolation would be required. Many ingenious, alternative ways of disposing of some, or all, of the nuclear waste inventory have been proposed, but geological disposal remains the only one that appears to offer safe, long-term disposal of all waste types. The report by the UK's Committee on Radioactive Waste Management (CoRWM 2006) provides a comprehensive analysis of alternatives. Nuclear nations in western Europe and elsewhere had also begun to plan for geological disposal during the late 1970s and 1980s. Most countries have interacted through the International Atomic Energy Authority, which has established safety requirements for the disposal of radioactive waste (IAEA 2011a) and guidance on how geological disposal facilities should be developed (IAEA 2011b). A history of ten national programs was published recently by the Nuclear Waste Technical Review Board (2015) and, in part, is summarized in the article by Metlay (2016 this issue).

By combining these articles on different repository types into a single issue of *Elements*, the reader has the opportunity to compare and reflect on the different strategies. The good news is that there are a number of very different strategies, some of which seem close to coming to fruition. The bad news is that, after 50 years of effort, there is no geological repository receiving highly radioactive waste. Countries with small nuclear programs, such as Sweden and Finland, have moved forward with recent approvals for the construction of repositories. In many countries with larger nuclear programs, such as the United States and Germany, the process appears to have stalled, although France continues to make progress. With this issue of *Elements*, we now have the opportunity to compare and evaluate the technical basis for the different strategies.

RADIOACTIVE WASTE TYPES

The types of waste intended for geological disposal in each country depend on both the nuclear fuel cycle adopted by that country and the national decisions about how to classify different radioactive materials. Waste types are discussed in more detail in the Box. For the purposes of this discussion, nuclear waste falls into three broad categories: 1) highly radioactive, heat-producing waste, mainly spent nuclear fuel and high-level waste from reprocessing of spent nuclear fuel; 2) less radioactive waste materials contaminated by long-lived nuclides, such as plutonium, which is generally termed "long-lived intermediate-level waste" in Europe or "transuranic waste" in the US; 3) low-level waste, mainly contaminated by short-lived radionuclides generated by medical procedures and reactor operations and suitable for shallow disposal. The first two categories are generally intended for deep, mined geological disposal.

From the perspective of geological disposal, the four critical issues for developing a repository are: 1) the radionuclide inventory; 2) how this inventory changes over time; 3) some sense of the geochemical mobility and radiotoxicity of the radionuclides; 4) the thermal output of the waste. The challenge for geoscientists is to understand how the evolving conditions of a repository affect the geochemistry and mobility of the radionuclide inventory that is also changing over time. The decay of one radionuclide often leads to the accumulation of another: for example, ^{239}Pu, which has a half-life of 24,100 years, decays to ^{235}U, which has a half-life of 700 million years.

Many proposed deep-mined geological repositories are for the disposal of heat-producing spent fuel or high-level waste. But within each of these waste types there are important variations. For spent nuclear fuel, the type of fuel, burn-up, and age since removal from the reactor all have an impact on the radionuclide inventory and the thermal output (Hedin 1997; Ewing 2015). For high-level radioactive waste, the type of chemical processing and the age of the waste are important. Reprocessing generally lowers the content of long-lived actinides, but leaves high concentrations of shorter-lived fission products, such as ^{137}Cs and ^{90}Sr, which have half-lives of approximately 30 years and a large thermal output which in turn determines how long the waste must be stored at the surface before disposal. These complexities mean that countries with a long history of developing nuclear power plants and weapons programs have a wide variety of waste that requires different treatments and handling. By contrast, countries with waste from a limited range of commercial nuclear power plants have much less diverse waste streams.

MULTIPLE BARRIER CONTAINMENT STRATEGIES

A major tenet of radioactive waste management is the concept of multiple barriers. This "belt and suspenders/braces" approach envisions a series of barriers, each with a capacity to prevent, or lower, the release of radionuclides to the environment. The concept reflects a prudent approach in the face of the huge uncertainties that result from projections of physical and chemical processes over large scales, both spatial (tens of kilometers) and temporal (hundreds of thousands of years). If one barrier is less effective than expected, other barriers will provide a margin of safety. The barriers are of two types: engineered and geological.

Engineered Barriers

Engineered barriers include the waste form, the waste package, and the surrounding backfill. Most of the engineered barriers are physical in that they delay the access of water to the waste package (e.g. the Ti-drip shields proposed at Yucca Mountain, Nevada, USA) or they delay the release of radionuclides from a breached canister (e.g. the bentonite buffer in the KBS-3 concept for granite repositories [see Hedin and Olsson 2016 this issue]). In some cases, the engineered barrier has a chemical function, affecting the geochemical environment around the waste. For example, MgO is emplaced with the transuranic waste in the bedded salt repository at the Waste Isolation Pilot Plant (WIPP) in New Mexico, and high-pH cement was developed in the UK to encapsulate intermediate-level waste. In both cases, the intention is to remove CO_2 and provide an environment in which the solubility of actinides is lowered.

Waste Form

Although the most common waste forms for highly radioactive materials are spent nuclear fuel and vitrified high-level waste, there are many other types of waste forms (Lutze and Ewing 1988; Weber and Ewing 2013). A previous issue of *Elements* (2006 v2n6, "The Nuclear Fuel Cycle: Environmental Aspects") has articles on spent nuclear fuel, borosilicate glass waste form, and crystalline ceramic waste forms. The properties of the waste form can be of critical importance for the simple reason that the waste form initially contains *all the radioactivity*. To the extent that the waste form is durable and the release of radioactivity is low, the remainder of the safety analysis is easier. One approach is to tailor the properties of the waste form to match either the composition of the waste stream or the geological conditions of the waste form in the repository. As an example, the UO_2 in spent nuclear fuel is unstable under oxidizing conditions, but much more stable under reducing conditions. Also, the corrosion rate of the borosilicate glass is slower when the silica concentration in the groundwater is higher, although other chemical components can lead to accelerated corrosion. The properties of the waste form change as the temperature and radiation field evolve with time (Ewing 2015).

Radiation damage produced mainly from the alpha-decay of actinides can lead to radiation-induced phase transformation from the crystalline to the amorphous state (Ewing and Weber 2010).

Waste Package

The waste form is typically placed in a metal canister, which may consist of steel, copper, or more advanced corrosion-resistant alloys. In many countries, the waste producers (i.e. utilities) are required to package the waste into standard packages that the repository is designed to accommodate. Some countries intend to follow the KBS-3 concept, developed in Sweden: here, spent fuel is placed in iron holders which are then encapsulated in copper containers, and these containers are then inserted into holes drilled into crystalline rock and lined with bentonite. Intermediate-level wastes are commonly encapsulated within stainless steel or iron canisters, with or without added cement.

Back-fill or overpack

In order to reduce water movement through a repository after closure, the remaining void space must be filled with a material compatible with the engineered and geological barriers. Bentonite backfill for heat-producing waste canisters has been investigated extensively. For larger waste volumes, host rock materials and cement are likely to be important. Repositories in salt will employ crushed salt, which will gradually be compacted as the excavated rooms close. Uniquely, the proposed repository at Yucca Mountain does not utilize a backfill (Swift and Bonano 2016 this issue).

Geological Barriers

Geological barriers exploit the properties of the rock and hydrologic system around the repository and are intended to extend the travel time for radionuclides to return to the biosphere. The movement of fluid through the rock is controlled by its hydraulic conductivity, which depends on matrix permeability and the presence or absence of fracture systems. Irrespective of the movement of groundwater, the mobility of the radionuclides in solution may be further retarded by dilution, sorption onto mineral surfaces, precipitation of secondary phases that contain radionuclides, and matrix diffusion.

All the proposed repository sites (to date) are located below the water table—with the exception of the Yucca Mountain site in Nevada where the proposed repository is in the unsaturated zone (Swift and Bonano 2016 this issue). The apparent "dryness" of the unsaturated zone is complicated by a high infiltration rate, the large volume of water that is held in the porosity, and rapid flow along fractures. For disposal below the water table, the host rock must have a low permeability to ensure that radionuclides released from containers over time do not reach the biosphere. Three types of host rock are widely recognized as having the potential to provide a sufficiently low permeability to meet this safety requirement: 1) strong rocks with very low porosity and very few fractures, including igneous, metamorphic, and certain types of sedimentary rocks (see Hedin and Olsson 2016 this issue); 2) relatively weak mudrocks or clays, which will not sustain open fractures for extended periods of time (Grambow 2016 this issue); 3) rock salt, which is also self-sealing (von Berlepsch and Haverkamp 2016 this issue). The rock must also have the necessary physical properties to construct a repository within it. In some cases, as in granite, high mechanical strength favors construction, but the presence of fracture systems may increase the hydraulic conductivity. In other cases, such as salt, a medium level of mechanical strength may be advantageous because plastic deformation and flow will seal the rock around the waste packages.

Thus, the petrophysical, geochemical, and hydrological properties of the geological formation have an important impact on the strategy that is adopted with the engineered barriers. Further, there are important interactions between the engineered and geological barriers. The geochemistry of the groundwater at the repository horizon may buffer its geochemical properties, which, in turn, will affect the rates of canister and waste-form corrosion. The physics and chemistry of the safety assessment is essentially an evaluation of how effective the barrier systems are going to be over time, a problem complicated by the coupled and evolving thermal, hydrologic, and geochemical conditions of the repository. The strategy for handling the heat of the spent nuclear fuel and high-level waste will also dictate the disposal strategy and the choice of barriers. Aged-waste in smaller packages can reduce the thermal pulse in the repository, which is important to avoid breakdown of clays if these are used in the backfill. On the other hand, larger packages and a larger thermal pulse might be used to drive water away from the surface of the waste package. Very high heat, however, can affect the condition of fuel cladding: this is a thin metal alloy wrapped around the fuel pins and in some concepts, is also considered a barrier. Also, the safety assessment has to consider regional aspects of the geology, such as seismicity, volcanism, and even climate change and glaciation.

The presence of natural resources in a region must also be considered because the exploration for and extraction of resources can compromise a geological repository.

REGULATORY FRAMEWORK

Once engineers have designed the repository and scientists have studied the performance of all of the processes that affect the barrier systems, then the performance of the system of engineered and geological barriers has to be placed into a regulatory framework. The critical parameters that are measures of the success of the repository strategy may vary. In the US, early regulations specified fractional release for the different engineered barriers as compared to the inventory at some time, say 1,000 years after emplacement. For geological barriers, minimum travel times were defined for the groundwater system. These early requirements lead to subsystem requirements on each barrier that were finally judged to be suboptimal, and the early regulation with subsystem requirements was replaced by a risk-based approach. The risk-based approach requires a total system performance analysis that probabilistically provides a quantitative estimate of the dose to a person or population at a certain place and time, usually extending to one million years (the so-called "compliance period"). Although one certainly wants to understand anticipated doses to the affected public, this calculation adds the exposure pathways in the biosphere to the risk calculation. The biosphere's complexity increases the uncertainty of the safety analysis.

While most countries extend their safety analysis to one million years, the level of the quantitative requirement varies. The Swedish KBS-3 concept is intended to provide containment of radioactivity within the engineered barriers until it has decayed to the level of the original uranium ore from which the waste was derived. Safety analyses may be based on quantitative determinative models early in the disposal period and then be replaced by more qualitative arguments for the very long periods involved. Thus, the safety analysis itself becomes a major subject of research (Ewing et al. 1999).

Nearly as important as the compliance period is the position of the compliance boundary, i.e. the point at which calculations are made to show the level of exposure to the public. The point of compliance can vary dramatically, even within a single country. In the United States, the compliance boundary for Waste Isolation Pilot Plant (WIPP) is a square, four miles on an edge around the repository. At Yucca Mountain, the compliance boundary was a long rectangle, the long axis parallel to the expected movement of groundwater, 17 miles in length. This has the effect of including the dilution and sorption processes along this transport path as part of the barrier systems of the repository. Of course, all of these calculations require some confidence in our understanding of where people will live and their daily habits far into the future.

PUBLIC ACCEPTANCE

Even when there is a strong scientific basis for the safety analysis and when a regulatory agency has declared that a geological repository is safe, public acceptance is key to success. Public acceptance does not come at the end of the process, but rather is earned throughout the process from the initial site selection through license approval. In this regard, the experience in different countries has been met with different levels of success (see Metlay 2016 this issue). There is, however, one aspect of the technical strategy that has a direct bearing on communication with the public: whether the analysis of different repository strategies is simple or complex. All things being equal, a robust strategy based on simple physical and chemical principles that are transparent when communicating with the public and politicians is probably more useful than a very sophisticated, probabilistic analysis that is less transparent. In fact, every safety analysis requires both a simple, accurate description and a more complex analysis. An important responsibility for scientists is to be sure that the simple analysis is not a "fig leaf" for the more complicated processes that are so common in geologic systems.

As guest editors, we hope that we have given you enough guidance so that as you read each article you can appreciate the waste disposal strategy and identify the role and efficacy of the different barrier systems. And now imagine … if you had the responsibility for developing a geological repository for nuclear waste, which strategy would you adopt?

REFERENCES

Bredehoeft JD, England AW, Stewart DB, Trask NJ, Winograd IJ (1978) Geologic Disposal of High-Level Radioactive Wastes—Earth-Science Perspectives. U.S. Geological Survey Circular 779, 15 pp

CoRWM (2006) Managing our Radioactive Waste Safely. Committee on Radioactive Waste Management Recommendations to Government. CoRWN Doc 700, 195 pp, https://www.gov.uk/government/uploads/system/uploads/attachment_data/file/

294118/700 - CoRWM July 2006 Recommendations to Government pdf.pdf, Accessed May 25, 2016

Ewing RC (ed) (2006) Nuclear Power: Environmental Impact. Elements 2: pp 321-392

Ewing RC (2015) Long-term storage of spent nuclear fuel. Nature Materials 14: 252-257

Ewing RC, Tierney MS, Konikow LF, Rechard RP (1999) Performance assessments of nuclear waste repositories: a dialogue on their value and limitations. Risk Analysis 19: 933-958

Ewing RC, Weber WJ (2010) Actinide waste forms and radiation effects. In: Morss LR, Edelstein NM, Fuger J (eds) The Chemistry of the Actinide and Transactinide Elements, Volume 6. Springer, New York, pp 3813-3888

Grambow 2016 this issue

Hedin A (1997) Spent Fuel – How Dangerous is It? SKB Technical Report TR-97-13: 60 pp

Hedin and Olsson 2016 this issue

IAEA (2011a) Disposal of Radioactive Waste. Specific Safety Requirements. International Atomic Energy Agency Safety Standards Series No. SSR-5, 83 pp

IAEA (2011b) Geological Disposal Facilities for Radioactive Waste. Specific Safety Guide. International Atomic Energy Agency Safety Standards Series No. SSG-14, 124 pp

Lutze W, Ewing RC (eds) (1988) Radioactive Waste Forms for the Future. North-Holland Physics Publishing, Amsterdam, Netherlands, 778 pp

Metlay (2016 this issue)

NAS–NRC (1957) Disposal of Radioactive Waste on Land. National Academy of Sciences–National Research Council report. National Academies Press, Washington, DC, 142 pp

NAS–NRC (1978) Geological Criteria for Repositories of High-Level Radioactive Waste. National Academy of Sciences–National Research Council report. National Academies Press, Washington, DC, 19 pp

Nuclear Waste Technical Review Board (2015) Designing a Process for Selecting a Site for a Deep-Mined, Geologic Repository for High-Level Radioactive Waste and Spent Nuclear Fuel: Overview and Summary. US Nuclear Waste Technical Review Board, 275 pp

Swift and Bonano 2016 this issue

von Berlepsch and Haverkamp 2016 this issue

Weber WJ, Ewing RC (2013) Ceramic waste forms for uranium and transuranium elements. In: Burns PC, Sigmon GE (eds) Uranium – Cradle to Grave. Mineralogical Association of Canada, Short Course Series 43: 317-336

BOX

WASTE TYPES: VOLUMES, MASSES, AND ACTIVITY THROUGH TIME

The definition of different waste types determines the strategy for handling them, e.g. shallow disposal versus deep geologic disposal. The largest volume of radioactive waste is low-level waste (LLW) and very-low-level waste (VLLW). How this is defined varies between countries, and the vast majority of these waste types will be disposed of in shallow, near-surface repositories, rather than by geological disposal.

The most radioactive materials are spent fuel (SF) or the high-level waste (HLW) that is derived from SF reprocessing. When spent fuel is removed from a reactor it contains fission products (mainly stable isotopes), plutonium, traces of other actinides, and uranium. If fuel is reprocessed, the fission products are present in the resulting high-level waste, which is most commonly vitrified for geological disposal. Some of the fission products are radioactive, have relatively short half-lives, and generate significant heat. These wastes have to be kept in interim storage for several decades while the isotopes ^{90}Sr and ^{137}Cs in particular undergo appreciable decay because they will continue to produce heat for hundreds of years. Spent fuel and HLW typically contain over 95% of the radioactivity in national radioactive waste inventories, despite making up only a few percent of the volume. Because the volumes are small, it is possible to package these types of heat-producing wastes in very resistant containers that will outlast most of the radioactivity, meaning that the waste does not interact with groundwater until after all but long-lived radionuclides remain. This technique is the one used in the Swedish KBS-3 concept.

The bulk of the waste volume for geological disposal consists of, in European parlance, long-lived intermediate-level waste (ILW), which includes scraps and waste generated during reactor operation, decommissioning, or reprocessing. For example, after the metal holders of fuel rods have been dismantled for reprocessing, scraps of spent fuel still adhere to them. This material is also designated for geological disposal and presents particular challenges because it is longer-lived than SF and HLW, yet cannot be packaged in such durable containers because of the much greater volumes.

FIGURES 1 and 2 show the projected wastes that the United Kingdom will send for geological disposal, assuming an additional generation of reactors is built, and that eventually all stockpiles of uranium (U) and plutonium (Pu) are treated as waste. FIGURE 1 shows the volumes of the package wastes and the masses of the waste. Clearly, ILW and U predominate, with relatively small amounts of the highly radioactive, heat-producing wastes (SF, HLW, and Pu). FIGURE 2 shows the contributions of the different waste types to the activity of the inventory at the projected time of opening of a geological repository in 2040 and onwards, with the area of the circles scaled to the total activity. The major source of radioactivity is spent fuel (much from reactors that have not yet been constructed), but eventually the contribution from uranium becomes important. The voluminous ILW is never a major contributor to the total activity, and when timescales reach hundreds of thousands of years, then uranium becomes an increasingly dominant source of activity because its long half-life means that it undergoes very little decay over this period.

Caption to Box Figures

FIGURE 1 Pie charts representing the relative proportions of the different types of radioactive materials in the UK's 2013 inventory of waste that has been designated for geological disposal by packaged volume and by conditioned mass. Abbreviations are as follows: U (uranium), LLW (low-level waste), ILW (intermediate-level waste), HLW (high-level waste), Pu (plutonium), SF (spent fuel).

FIGURE 2 Pie charts to show the contributions of different waste types to the projected activity of the UK's 2013 inventory for geological disposal. Projections are shown for the times of opening and closure of a geological disposal facility and for times of 2,000 years and 200,000 years after closure. The last two pie charts are shown in expanded form so that the breakdown by waste type can be seen. Abbreviations as in Figure 1.

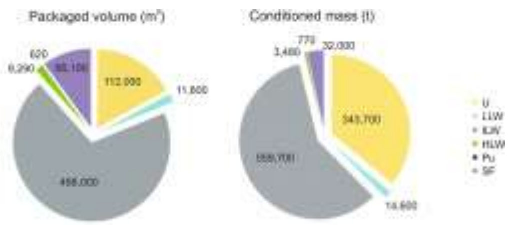

Figure 1

Figure 2