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Specialized Disposal Sites for Different Reprocessing Plant Wastes

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ABSTRACT

Once-through fuel cycles directly dispose of one primary waste form: spent nuclear fuel (SNF). In contrast, the reprocessed SNF yields multiple waste streams with different chemical, physical, and radionuclide characteristics. These different characteristics of each waste stream imply that there are potential cost and performance benefits to developing different disposal sites that best match the disposal requirements of each waste stream. Disposal sites as defined herein may be located in different geologies or in a single repository containing multiple sections, each with different design characteristics. In the context of the Global Nuclear Energy Partnership, the long-term potential may exist for a global waste management system that employs multiple disposal facilities in which the physical, chemical, and radiological characteristics of a particular waste stream can be matched to the waste isolation characteristics of a specific disposal site. The paper describes the results of a series of studies on disposal options for specific wastes and the potential for a waste management system that better couples various reprocessing plant wastes with disposal facilities.

INTRODUCTION

Repository design, performance, and costs are controlled by four factors associated with each waste stream: (1) the physical characteristics, (1) the decay heat, and (3) the potential for specific radionuclides to escape the repository, and (4) radiation levels. To understand the disposal options for various wastes, the waste streams and their characteristics must be defined.

Reprocessing plants produce multiple waste streams that may include (1) uranium; (2) plutonium; (3) minor actinides (MA); (4) high-heat radionuclides (HHR) containing ^{137}Cs and ^{90}Sr with their decay products; (5) volatiles and their subsequent decay products that would be released during reprocessing operations including inert gases (He, Ne, Ar, Kr, Xe, and Rn), halogens (F, Cl, Br, I), hydrogen, nitrogen, and carbon; (6) the remaining radionuclides that are very-low heat radionuclides (VLHRs), and (7) structural materials such as zirconium clad from light-water reactor fuel. Table I shows these categories and their heat generation rates as a function of time [1].

While SNF contains hundreds of radionuclides, in any repository only a few radionuclides determine the risk to the public and thus control repository design. For example, Fig. 1 shows the expected radiation doses to the public [2] from the proposed Yucca Mountain (YM) repository versus time. The analysis indicates that ^{99}Tc and ^{129}I control the maximum dose to the public over time. If SNF is reprocessed and these radionuclides are naturally or deliberately separated from the other wastes, serious consideration should be given to developing methods that can better isolate these radionuclides from the biosphere. If these radionuclides can be better isolated from the environment, the risk to the public is decreased and repository capacity (based on allowable radionuclide releases from the repository) can be increased.

Table I. Streams from Processing 1 Metric Ton Initial Heavy Metal of 40,000 MWd/t of Pressurized Water Reactor SNF

	SNF	U/Pu	HHRs	VLHR	MAs	LHRs	Volatiles
Mass[a] (g)	1.427×10^{6}	9.576×10^{5}	4.132×10^{3}	3.030×10^{4}	1.192×10^{3}	7.206×10^{3}	2.918×10^{5}
Decay Heat[b] (watts)							
At 10 years	1.443×10^{3}	1.851×10^{2}	1.024×10^{3}	6.359×10^{1}	1.132×10^{2}	8.900×10^{0}	4.799×10^{1}
At 20 years	1.096×10^{3}	2.113×10^{2}	7.554×10^{2}	2.239×10^{1}	8.989×10^{1}	4.663×10^{0}	1.284×10^{1}
At 50 years	6.578×10^{2}	2.276×10^{2}	3.726×10^{2}	1.972×10^{0}	5.457×10^{1}	6.708×10^{-1}	3.051×10^{-1}
At 100 years	3.555×10^{2}	2.014×10^{2}	1.154×10^{2}	8.611×10^{-2}	3.858×10^{1}	2.668×10^{-2}	4.551×10^{-2}
At 1000 years	6.308×10^{1}	5.395×10^{1}	1.818×10^{-4}	2.353×10^{-2}	9.097×10^{0}	1.739×10^{-4}	1.531×10^{-2}

[a]SNF includes 1.334×10^{5} g oxygen. Component streams exclude oxygen.
[b]Time measured from reactor discharge. Separations assumed to occur 5 years after reactor discharge

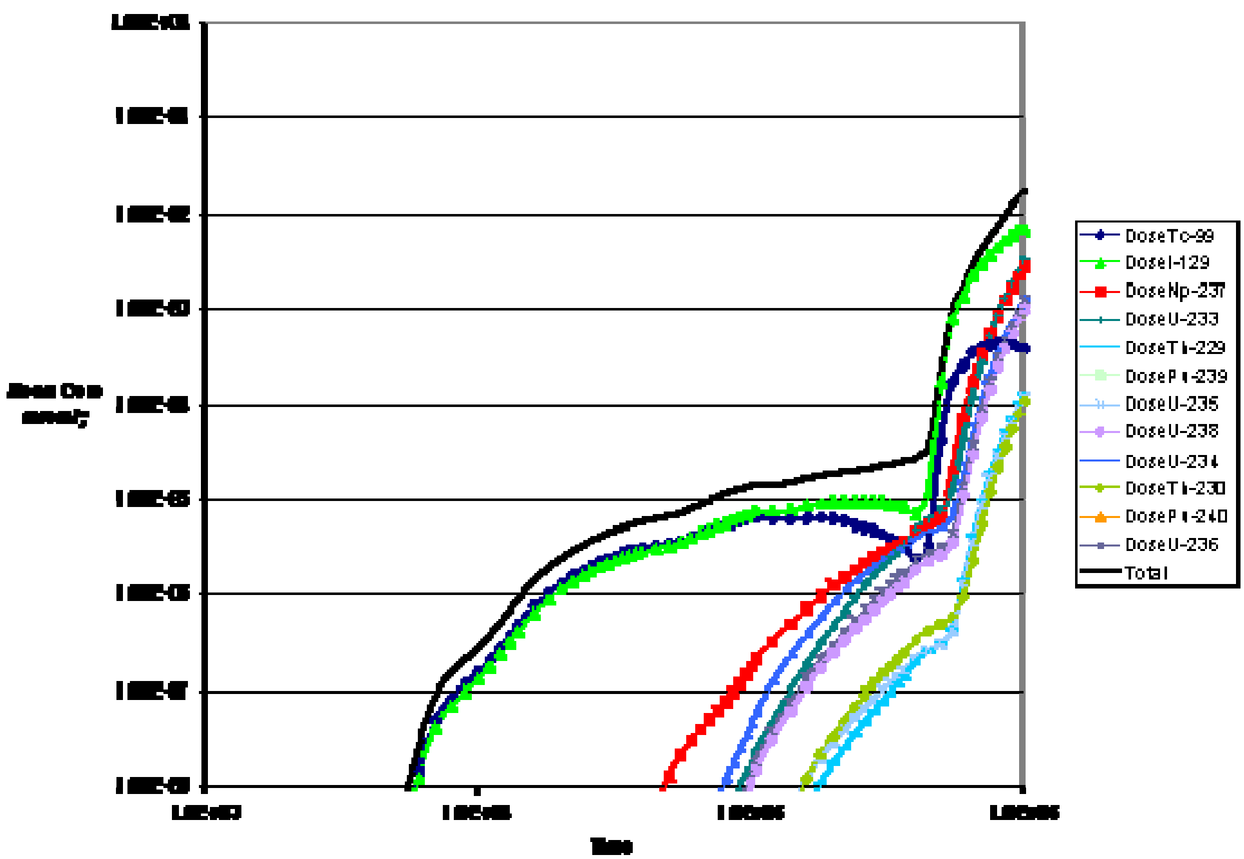

Figure 1. Expected radiation exposure from YM versus time.

HIGH-HEAT WASTES

The high cost of geological repositories is primarily a consequence of the decay heat generated by the wastes [1]. To prevent overheating of the waste packages (WPs) and the geology, the wastes must be distributed over a wide area. This, in turn, requires kilometers of tunnels and thousands of expensive WPs. If there were no decay heat, a repository would consist of a couple of silos, such as modified variants of the intermediate-level-waste high-activity silos in the Swedish Final Repository for Radioactive Operational Waste (SFR). The SFR silos [3] were excavated in granite under the Baltic Sea with access by tunnel. Each silo is 50 m high and 25 m in diameter. A thick bentonite clay barrier surrounds the silo and fills the space between the rock cavern and the silo. The clay barrier serves as (1) a barrier to water migration and (2) a mechanism to retard radionuclide migration. The wastes are placed in the silo and cemented in place using a special cement grout. Each silo is a massive, low-cost WP.

The three classes of heat-generating radionuclides are plutonium, the MAs, and the HHRs. Plutonium is a product of reprocessing and thus is recycled. The MAs may or may not be recycled. The HHRs are a waste stream with a small mass (4.1 kg per ton of SNF) that after 10 years from reactor discharge accounts for 71% of the decay heat, 89% of the gamma rays, and 99% of the ingestion hazard of SNF. The half-lives are ~30 years; thus, their mass decreases by a factor of 10 each century. Three strategies for HHR geological disposal have been identified.

Vented Repository. All repository designs have maximum allowable temperatures to avoid degradation of barriers to radionuclide migration such as the waste form, WP, and geology. These temperature limits, in turn, determine the maximum allowable decay heat per WP, the spacing of the WPs, and the spacing of the disposal drifts. However, these limits are set for a closed repository where decay heat removal is by conduction of decay heat to the surface. If the repository is designed to allow active ventilation of the WPs for some period of time, this decay heat can be removed. If the repository remains open and is ventilated for 200 years, the decay heat from the HHRs is insignificant and therefore HHRs do not control repository design. As the repository ventilation time increases [4], the repository size decreases and the number of WPs decrease because larger WPs that are more cost-effective per unit volume can be used.

High-Heat Repository. If the HHRs are separated, the HHR capsules may be disposed of in a small high-heat repository. One option for the YM site [1] is to mine a tunnel through the middle of the HHR disposal zone. Horizontal boreholes (10- to 15-cm diam.) many hundreds of meters long are drilled into the rock from this tunnel and filled with small-diameter HHR capsules. This design avoids construction of large disposal drifts. The HHR section of the repository is designed as an “extended-dry” repository in unsaturated rock where the local rock temperature is driven by the decay heat above the boiling point of water for thousands of years. The expected failure mode of a repository is capsule failure, ground water dissolution of radionuclides or formation of transportable colloids, transport of radionuclides to the open environment by groundwater, and inhalation or ingestion of the radionuclides by people. If the rock temperature is above the boiling point of water, there can be no groundwater flow and, therefore, no transport of radionuclides to the open environment. The HHRs decay before the section of the repository cools below the boiling point of water. Because ^{135}Cs is the only long-lived radionuclide in this system, only a long-term performance assessment of this isotope needs to be considered. With a single radionuclide, such a performance assessment becomes tractable for a repository section that goes through a severe temperature transient. Because of the high heat loading and allowable temperatures, the HHR zone is very small relative to traditional repositories.

Salt Diver. The salt-diver repository uses the high-heat generation rates of HHR capsules to allow disposal at depths up to 10,000 m underground in salt domes. The HHRs are packaged into moderately large containers (salt divers) that are placed in a salt dome. The high-density salt-diver heat source sinks by heating the salt under the WP until the salt becomes plastic. Salt melts at 800°C but is plastic at much lower temperatures. The salt diver then sinks to the bottom of the salt dome. The long-term tendency of hot WPs to sink in salt is one of the design considerations of salt repositories. Launching would require placing the salt diver on the salt floor or in a hole in the salt floor of the facility. Because the same launch site could be used repeatedly, no excavation of kilometers of tunnels in the salt would be needed.

This may be the lowest-risk, lowest-cost disposal option for HHRs. To a first-order approximation, waste isolation improves with depth. There is no realistic potential for accidental human intrusion in the future. The safety case is simple. A typical salt dome contains cubic kilometers of salt. The time to dissolve a significant fraction of the salt far exceeds the time it would take for the ^{137}Cs and ^{90}Sr to decay. Furthermore, the HHRs are at the bottom of the salt dome. As salt dissolves, insoluble materials will fill the void space and thus lower the salt dissolution rate.

LOW-HEAT WASTES

The controlling design requirement is to isolate the LHRs from the environment for a very long time. The optimum design (assuming very low heat-generation rates) is a single, large sphere buried deep underground. The engineered version is a large WP or a build-in-place large cylinder such as the SFR silos. The primary radionuclide release mechanism is (a) dissolution of radionuclides in groundwater and (b) the transport of that groundwater to the open environment. Large WPs minimize radionuclide release by two mechanisms.

- *Mass transfer*. Various barriers can be placed around the waste to slow the flow of water through the waste. With a smaller surface-to-volume ratio, fewer economic constraints are placed on the design of such barriers; consequently, higher performance barriers may be used.

- *Solubility limits*. The release of radionuclides from a WP after its failure is proportional to the groundwater flow through the waste and the solubility limits of the radionuclides in groundwater. Let us consider a radionuclide A that will dissolve in groundwater up to its solubility limit. In one scenario, a fixed quantity of radionuclide A is spread over 25 m^2 of WPs. In a second scenario, the same quantity of radionuclide A is spread over 1 m^2 of WPs. Assuming a uniform groundwater flow, the flow through this 1-m^2 area is one twenty-fifth that of the flow through the 25-m^2 area. Because the flow of water through the waste is reduced by a factor of 25, the release rate is reduced by a factor of 25. Larger WPs with smaller cross-sectional areas per unit of waste lower the total repository radionuclide release rates.

Various studies [5, 6] have modeled and quantified these benefits. Furthermore, these design principles are used in the design of various low- and intermediate-level waste disposal facilities that have low heat-generation rates.

BOREHOLE

Deep boreholes can be drilled to ~10,000 m for the disposal of wastes [7–10]. The technology offers extreme isolation, which makes it potentially very attractive for very-long-lived, escape-prone wastes such as ^{99}Tc and ^{129}I. Cost factors may limit the use of this technology to only these small volume wastes, but the technology is advancing rapidly because of the need to drill deeper oil, natural gas, and geothermal wells.

The potential for extreme waste isolation as compared to traditional repositories is based on several factors. The distance to the biosphere is large and regions are accessed where nothing has happened on geologic timeframes. Rock is chosen where the groundwater is as old as the rock itself and has not moved into or out of the host rock since they were laid down together. In most parts of the world, the groundwater goes from fresh water to seawater to highly saline waters that have significantly higher densities. Because of these higher densities, these groundwaters can not move unless there is excessive decay heat to overcome this natural stability. These mechanisms make it impossible for radionuclides to escape by dissolution and be transported in groundwater.

SEABED

Wastes can be disposed of under the ocean [11, 12] in mined tunnels, boreholes, or other structures. Seabed disposal can provide extreme isolation relative to most other methods of disposal. Two methods are used to ensure safety from any hazardous material: (1) geological disposal and (2) extreme dilution. Seabed disposal uses both of these methods. Theoretical analysis indicates that if radionuclides were randomly disposed of in the earth’s crust, the fraction that would enter the food chain each year would be between 10^{-11} and 10^{-12}. For radionuclides in the ocean, the fraction that would enter the food chain is 10^{-9} to 10^{-10}. Only this method uses ocean dilution as the backup mechanism.

This option is potentially most attractive for ^{129}I and other radionuclides that are normally in chemical forms that are highly soluble in groundwater. This isotope is a primary contributor to repository risk because continental locations have little iodine and the human body concentrates the iodine it obtains in the thyroid with remarkable efficiency. However, the ocean has massive quantities of natural nonradioactive iodine. If ^{129}I leaks from a disposal site under the ocean, it is diluted by massive quantities of nonradioactive iodine.

REDUCING AND OXIDIZING REPOSITORIES

Local chemistry determines which radionuclides may migrate over time. If multiple disposal sites are available, the option exists to choose the disposal site with the geochemistry that provides the best isolation for each waste stream. This is not an option for direct disposal of SNF which is a single waste form. At YM, an oxidizing geochemical environment, the four most important radionuclides [2] are ^{129}I, ^{99}Tc, ^{237}Np, and ^{233}U. In contrast, in the proposed Swiss repository which has chemically reducing conditions, the four most important radionuclides [13] are ^{14}C, ^{129}I, ^{36}Cl, and ^{79}Se. Except for ^{129}I, a different set of radionuclides determines the long-term potential dose to the public.

GASEOUS WASTES

Gaseous wastes such as krypton present special challenges. While krypton can be stored in cylinders until the ^{85}Kr decays ($T_{1/2}$ = 10 years), there are the risks of release from high-pressure gas cylinders. Krypton can be incorporated into solids, but the technologies are difficult and expensive. One geological strategy has been identified for disposal of the gas—dissolution into saline water in the same deep geological structures being proposed for the sequestration of carbon dioxide. This is a variation of an earlier disposal option developed in Germany in which the gas was compressed into cylinders with one-way valves. The cylinders are dropped into the deep ocean where water enters the cylinders at depth and the krypton then dissolves into the seawater in the cylinder. The ocean disposal option has multiple levels of protection. The cylinder lifetime exceeds the lifetime of the radioactive ^{85}Kr, and the rate of ocean circulation ensures decay of any ^{85}Kr long before the deep ocean water approaches the surface of the ocean and could release the ^{85}Kr to the atmosphere.

CONCLUSIONS

Unlike direct disposal of SNF, the reprocessing of SNF creates multiple waste streams with different physical, decay heat, and radionuclide characteristics. These multiple waste streams pose the possibility of using different disposal sites within the same geology or within different geologies to dispose of specific wastes with potential reductions in risk and cost. There are licensing and other constraints, particularly for saltdiver, borehole, and seabed options that have not been fully developed. Although there are complex economic, institutional, and technical trade-offs to be considered, such strategies may offer significant benefits to a global nuclear energy system.

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