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IMPACTS OF NUCLEAR FUEL CYCLE CHOICES ON PERMANENT DISPOSAL OF HIGH-ACTIVITY RADIOACTIVE WASTES

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Abstract

All options for generating power from nuclear energy generate radioactive waste products that will require permanent isolation from the biosphere. Choices made regarding nuclear fuel cycle options, including decisions for recovery and re-use of fissile material from irradiated fuel, have the potential to affect the waste stream characteristics such as mass, volume, radioactivity, and thermal power, but no options eliminate the need for robust isolation of wastes. Decades of experience has produced an international consensus that deep geological disposal is the preferred method for achieving permanent disposal. The paper reviews published results of safety assessments for deep geologic disposal concepts that have been proposed in the United States, Sweden, France, Switzerland, and other nations to provide insight into the waste form aspects that most affect the long-term performance of repository systems. Disposal concepts considered include geologic repositories in multiple rock types in both saturated and unsaturated environments. Additionally this work evaluates how repository performance may be affected by hypothetical waste form modifications from changes in fuel cycle choices.

1. INTRODUCTION

The recognition that deep geological disposal is the preferred option for achieving safe isolation of high-activity radioactive wastes dates from the 1950s[1], and many nations began researching specific disposal options in the 1980s. Mined repositories are in operation for some categories of transuranic and intermediate-level waste[2,3] and a repository for spent nuclear is under construction in Finland[4]. Progress toward facility licensing has been slow elsewhere in the world, however[5].

This paper reviews published safety assessment results for five different disposal concepts: mined repositories in granite[6.4.7], argillite[8,9], salt[2,10,11], volcanic tuff[12], and deep borehole disposal in crystalline rock[13,14]. This work also provides insights on how specific changes to the waste form that might result from alternative fuel cycle choices might affect long-term performance of each concept. Published analyses indicate that all five concepts have the potential to meet regulatory requirements and provide robust long-term isolation for the existing waste forms from the existing fuel cycles in each program.

Hypothetical modifications to waste forms requiring deep geologic disposal that could result from alternative fuel cycles and that are considered here include:

- Reduction in the radionuclide inventory associated with recovery and re-use of fissile isotopes
- Reduction in the radionuclide inventory associated with additional partitioning and transmutation of radioisotopes remaining after recovery and re-use of fissile isotopes
- Reduction in the volume of waste associated with recovery and re-use of fissile isotopes
- Reduction in the thermal power of the waste associated with recovery and re-use of fissile isotopes
- Increases in the durability of the waste form in the repository environment resulting from further treatment of the wastes
- Increases in the durability of spent nuclear fuel in the repository environment resulting from alternative fuel cycle choices.

2. BACKGROUND ON THE DISPOSAL CONCEPTS CONSIDERED IN THIS PAPER

The Radionuclide Inventory Requiring Geologic Disposal. Figure 1 shows the time-dependent radioactive content (activity) of typical light-water reactor fuel from the US following irradiation[12]. This example provides a useful representation of the radionuclides that require long-term isolation from any fission-based fuel cycle that does not include recovery and re-use of fissile material. At early times the disposal inventory is dominated by the relatively short-lived fission products Sr-90 and Cs-137. As these isotopes decay over the first few hundred years, the total amount of radioactivity becomes dominated by the transuranic radionuclides Am-241, Pu-240, and Pu-239. After several hundred thousand years, the long-lived fission product Tc-99 becomes the dominant contributor to the total inventory, until the system becomes dominated by Np-237, Pu-242, and long-lived isotopes of U and Th. These radionuclides are not necessarily the most important contributors to estimates of long-term releases from repositories because the mobility (and immobility) of specific radionuclides within each disposal concept is a key control on long-term releases.

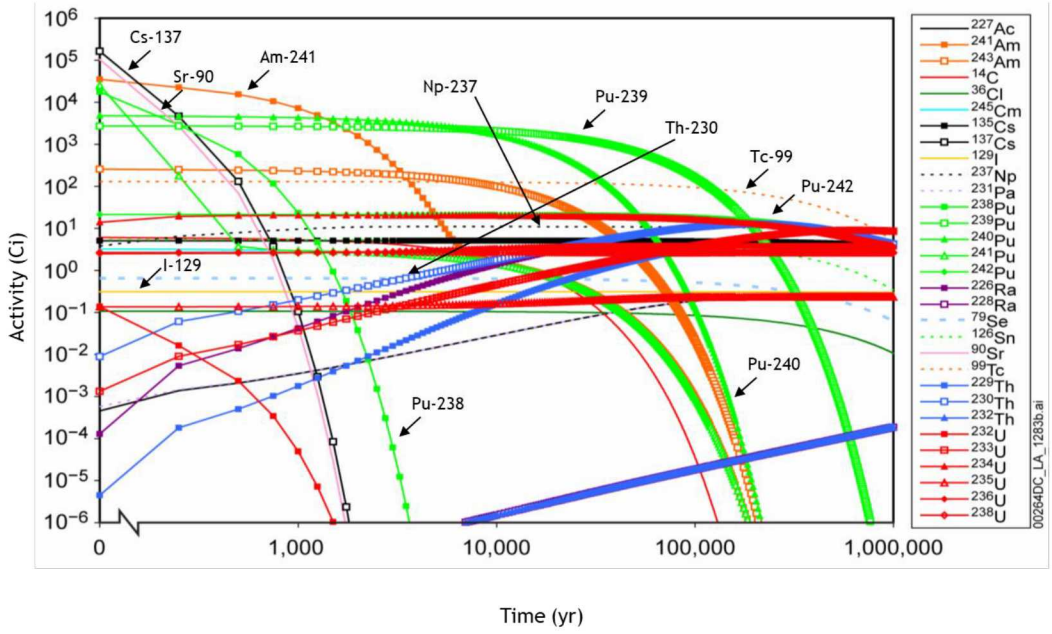

Figure 1. Example of radioactive decay/ingrowth in irradiated spent nuclear fuel. Inventory activity is shown for a single representative waste package in the proposed Yucca Mountain repository, USA. Time is shown logarithmically in years after 2117. Source: Ref. 12, Figure 2.3.7-11.

Mined Repositories in Granitic or Crystalline Rock. Published analyses of the proposed repositories at Forsmark in Sweden[6,15], Olkiluoto in Finland[4] and a generic site in Canada[7] provide representative examples of disposal in a mined repository in granite or granitic crystalline rock. The concept calls for emplacement of spent fuel in copper canisters in holes drilled in the floor or walls of a mined facility at a depth of several hundreds of meters in granitic crystalline rock. Groundwater at that depth is reducing, and primary barriers providing isolation of the waste include the low dissolution rate of uranium oxide (the primary component of spent fuel) in reducing groundwater, the stability of metallic copper in reducing groundwater, and the capability of a bentonite clay buffer emplaced around each canister to prevent advective groundwater flow in undetected fractures in the granite from reaching the canisters. When waste packages fail, radionuclide sorption in the bentonite and radionuclide precipitation in the reducing groundwater will lower the magnitude of releases that may reach the biosphere.

Mined Repositories in Volcanic Tuff. The DOE's 2008 license application for the proposed repository at Yucca Mountain in Nevada, USA[12,16] provides the only example of disposal in volcanic tuff. The concept calls for emplacement of both spent fuel and high-level radioactive waste (in the form of borosilicate glass) in corrosion resistant waste packages placed end-to-end in mined tunnels. The proposed facility is 200 to 300 meters below

the land surface and, because of topographic relief and the aridity of the surrounding region, is also more than 200 meters above the water table in an unsaturated and oxidizing environment. Primary barriers providing isolation include the low volume of water flow through the unsaturated rock and the long-life expectancy of the waste package (Alloy-22, composed of nickel with high concentrations of molybdenum and chromium and chosen for its high corrosion resistance in oxidizing environments) and the overlying titanium drip shield that (while intact) will prevent seepage water from contacting the waste package surface. When waste packages fail, radionuclide sorption on corrosion products and mineral phases along the transport pathway will reduce the magnitude of releases that may reach the biosphere.

Mined Repositories in Argillite. Published analyses from the Belgian, Swiss, and French programs[17,8,9,18] provide examples of mined repositories in clay-rich rocks. Specific details of rock properties and facility design differ among the three examples, but in each case the chemically reducing conditions in the repository and the lack of advective flow in the low permeability host rock contribute to the long-term isolation of the waste. When waste packages fail, mobility of radionuclides will be limited by precipitation in the reducing groundwater, sorption on clay minerals in the host rock, and the slow rate of diffusive transport through the host rock.

Mined Repositories in Salt. The US is currently disposing of intermediate-level transuranic waste in bedded salt at the Waste Isolation Pilot Plant (WIPP)[2], and Germany has both disposed of intermediate and low-level radioactive wastes in domal salt at Morsleben and investigated possible disposal of spent fuel and high-level radioactive waste in domal salt at Gorleben[11]. Details regarding potential release pathways and the amount of brine that might contact the waste differ between bedded and domal settings, but in all cases, isolation in salt relies primarily on the extremely low permeability of intact salt (primarily halite), which precludes advective transport of radionuclides away from the repository. Observations made here are also based in part on analysis in the US of the long-term performance of a generic bedded salt repository[10].

Deep Borehole Disposal in Crystalline Basement Rock. No national programs are currently pursuing the deep borehole disposal option (3 to 5 km deep disposal), but multiple investigations over the past twenty years have suggested that it may be a viable option for relatively small volume waste forms with physical dimensions suitable for emplacement in holes drilled from the land surface[13,14]. Attributes of the concept that contribute to long-term isolation of the waste include the anticipated conditions at increasing depths: reducing chemistry; decreasing permeability, including the low frequency of open fractures in crystalline (granitic or metamorphic) rocks below 2 to 3 km; and increasing fluid salinity and density, which counters thermally driven upward flow; as well as the extremely long diffusive transport path through the borehole seal system. Published performance assessment analyses conducted in the US[13,14] provide the basis for the observations below on how changes to waste form properties may impact deep borehole repository performance.

3. IMPACTS OF CHANGES TO WASTE FORM PROPERTIES ON DISPOSAL SYSTEM PERFORMANCE

Reduction in the radionuclide inventory associated with recovery and re-use of fissile isotopes. Recovery and re-use of fissile radionuclides from spent fuel will directly reduce the inventory of those radionuclides, per kW of electricity generated, in the waste for geologic disposal. However, this inventory change does not necessarily produce a proportional increase in the long-term safety of the proposed disposal systems. In the absence of disruptions that directly expose waste to the biosphere (such as human intrusion), estimates of the long-term performance of disposal systems are dominated by the most mobile radionuclides, rather than those that contribute the most to total radioactivity. As shown in Figure 1, the radioactivity of typical spent nuclear fuel will be dominated for most of the next several hundred thousand years by isotopes of Pu. However, Pu and other actinide elements have limited mobility in chemically reducing environments, and published safety assessments for most disposal concepts show essentially zero direct contribution to risk (in terms of estimated dose) from Pu, U, and other radionuclides proposed to be removed. Risk in these concepts comes instead from long-lived fission and activation products, specifically I-129 and to a lesser extent Se-79 and Cl-36, that are mobile in essentially all geochemical environments.

In contrast, the proposed repositories at Forsmark[6] and Yucca Mountain[12] provide exceptions to the observation that risk is dominated by I-129. Forsmark has reducing geochemical conditions and strong sorption of actinides on the bentonite buffer, and shows no direct contribution to dose from isotopes of Pu or U. The relatively short-lived Ra-226 ($t_{1/2}$ = 1600 years), however, shows up as the primary contributor to dose, exceeding the contribution from I-129 by roughly a factor of 5 [Ref. 6, Figure 13-18]. This result is consistent with the chemical mobility of Ra in reducing environments and its continuous ingrowth from immobile Th-230, coupled with site-specific models that allow for relatively rapid transport from the repository to the biosphere in fractures that directly intersect the waste emplacement regions. Comparable Ra-226 releases are not observed in other disposal concepts in reducing environments for which long transport times allow substantial decay of Ra-226, (including in crystalline rock where fractures do not directly intersect the waste emplacement region[7]).

Dose estimates for Yucca Mountain, shown in Figure 2, show dominant contributions from Pu-238 and Pu-242 throughout the million-year regulatory period, consistent with the relatively higher mobility of Pu (and other actinides) in the unsaturated oxidizing repository environment and oxidizing groundwater transport pathways. Other significant contributions at one million years come from Np-237; Ra-226, which in this case is generated by decay of mobile U-234 and Th-230 throughout the transport pathway; and I-129, which because of its high mobility contributes approximately 1/10th of the total dose at one million years despite contributing less than 1/100th of the radioactivity inventory available for transport at one million years (Figure 1).

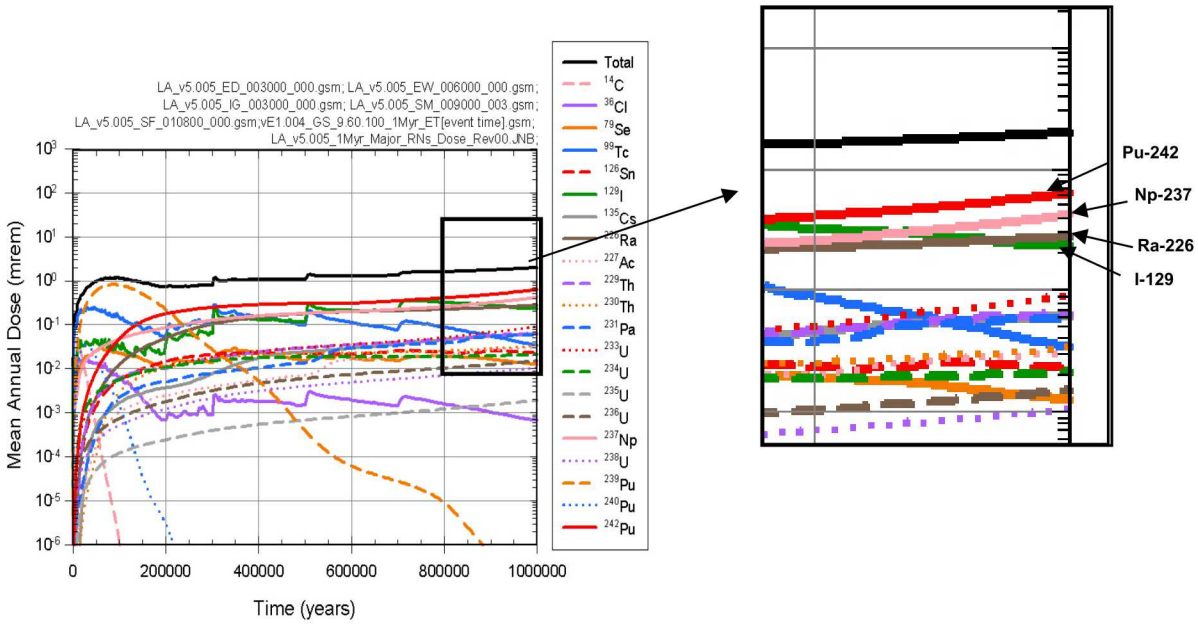

Figure 2. Estimated mean contributions from individual radionuclides to total mean annual dose resulting from the disposal of spent nuclear fuel and high-level glass waste in tuff [From Ref. 19, adapted from Ref. 12, Figure 2.4-20b].

In summary, radionuclide inventory reductions due to recovery and reuse of fissile isotopes of Pu and U can reasonably be expected to have no effect on estimates of the long-term performance of mined repositories in argillite or salt, or in crystalline rock concepts, mined or drilled, in which transmissive fractures do not directly intersect the waste emplacement region. In each case, reducing chemical conditions immobilize actinides and long transport times prevent all but long-lived mobile species, dominated by 1-129, from reaching the biosphere in significant quantities. For mined repositories in granitic or crystalline rock where relatively rapid transport to the biosphere may occur in fractures (e.g., Forsmark), removing the actinides that decay to create mobile Ra-226 would have a potential to reduce the estimated total dose by perhaps a factor of 5, at which point I-129 would become the primary dose contributor. Similarly, removing all actinides from the inventory of a repository in oxidizing conditions (i.e., Yucca Mountain) could decrease the estimated total dose by perhaps a factor of ten

before I-129 becomes the primary contributor. It should be noted, however, that the currently estimated doses for both Forsmark[6] and Yucca Mountain[12], which include these fissile isotopes, are well below regulatory limits such that there is no reason to suggest they would need to be reduced further to meet safety requirements.

Reduction in the radionuclide inventory associated with additional partitioning and transmutation of radioisotopes remaining after recovery and re-use of fissile isotopes. Nuclear fuel cycles have been proposed that will also remove minor actinides (specifically, Am, Np, and Cm) from the spent fuel in addition to U and Pu. As shown in the discussion above, and noted previously by multiple researchers [e.g., Ref. 20-23], reductions in the inventory of radionuclides of Am, Cm, and Np will have no perceptible effects on the estimates of total dose from disposal concepts that include reducing conditions and long transport times between the repository and the biosphere. Estimated doses from disposal concepts analogous to Yucca Mountain could be reduced by a small amount if Np-237 and its decay parent Am-241 were removed from the inventory before disposal, but, as noted in the previous section, the effect is limited to roughly a factor of ten before I-129 becomes the dominant contributor. In addition, various approaches have been proposed to transmute I-129 and other fission products into stable isotopes [e.g., Ref. 24-25]. Transmutation of fission products would clearly impact dose estimates from repositories if they could be implemented cost-effectively at industrial scales, but at this time there is little evidence to suggest that these techniques are realistic options for the future.

Reduction in the volume of waste and thermal power associated with recovery and re-use of fissile isotopes. Waste volume and thermal power are addressed together because, all other factors being held constant, the two properties are inversely correlated. Reductions in volume, unless they are accompanied by the separation and removal of heat-generating radionuclides, increase the thermal power per unit volume of waste. Decreasing waste volume has the potential to decrease the size of the repository and therefore decrease disposal costs, but increases in thermal power of the waste could counter that effect by requiring greater spacing between waste packages to meet repository design temperature constraints. Removal of heat-generating fission products and minor actinides from the waste stream has the potential to reduce waste volume without increasing thermal power, but there are multiple approaches to keep peak post-closure repository temperatures below a specified value that do not include partitioning and transmuting heat-generating radionuclides. For example, waste can be aged before disposal, the repository can be ventilated after waste is emplaced, waste package size can be decreased, and waste package spacing can be increased. Each of these approaches has been proposed in one form or another in published repository design concepts, and thermal constraints do not appear to limit implementation of any of the major disposal concepts for waste forms with a broad range of thermal output.

Although disposing of a wide range of thermal power waste is feasible, separating heat-generating radionuclides can result in substantial reductions in the required total excavated disposal volume. Modeling studies (e.g., Ref. 20) have evaluated variations in the thermal power of the waste and the spacing of waste packages while holding all other aspects of repository design and operations constant. As discussed by Ref. 19, the results suggest that the waste from a full-recycle fuel cycle (i.e., including separation of minor actinides) can meet the same temperature constraints for clay and granite repository concepts using only 30% to 40% of the disposal gallery length needed for disposing of waste from an equivalent electric-power-generating open fuel cycle [Ref. 20, table 7.1]. Doubling the aging time (to 100 years) for waste in which short-lived fission products are the dominant heat sources leads to further reduction in a hypothetical clay repository to approximately 8% of the original disposal volume [Ref. 20, Section 7.1.3].

For all disposal concepts, waste volume and thermal power considerations are probably best thought of as topics to be addressed through engineering design and cost optimization evaluations, and not as fundamental safety issues for disposal. Estimates of long-term dose from published safety assessments[6,7,8,9,12] meet existing regulatory requirements for waste forms from an open fuel cycle. Alternative waste forms may allow more efficient use of repository space or provide suitable geometries for small-diameter cylindrical waste packages for deep borehole disposal.

Increases in the durability of the waste form in the repository environment. Impacts of waste form durability may be evaluated for disposal of various forms of spent fuel without treatment (e.g., conventional light-

water reactor uranium oxide spent fuel[12], TRISO particle spent fuel[26]) and vitrified waste from reprocessed spent fuel (e.g., borosilicate glass). In all disposal concepts, radionuclide releases only occur once the waste form begins to degrade, and those releases then depend on the waste form degradation rate. Increasing waste form durability has been proposed as a means for improving overall repository performance[27], but because many other factors affect the timing and magnitude of the radionuclide source-term and radionuclide migration to the biosphere, the impacts of waste-form durability (i.e., lifetime) need to be evaluated in the context of the full disposal system. Other potentially significant factors include: water flux to/through engineered barriers containing the waste form; degradation rates of the engineered barriers; water chemistry contacting engineered barriers and the waste form; and radionuclide transport properties through engineered and natural barriers. Any of these factors may dominate overall performance of the repository if it controls the dominant radionuclides contributing to estimated dose, but in many cases disparate factors contribute to the dominant radionuclides contributing to the estimated dose. Therefore, it is not always clear whether improved performance of an individual aspect will translate into meaningful improvements of overall disposal system performance. For example, increasing the durability of a waste form may have little impact on the magnitude of the estimated peak dose if for example (a) the dominant radionuclide is solubility limited, or (b) the waste form lifetime is still relatively short compared the transport time to the biosphere.

As discussed by Ref. 19, the French safety assessment for an argillite disposal system[9] provides an example in which overall performance is relatively insensitive to waste-form lifetime because modeled releases are largely controlled by the slow rate of diffusive transport through the geosphere. For analyses assuming direct disposal of spent fuel, radionuclides were assumed to be released from the fuel over approximately 50,000 years [Ref. 9, p. 222]. Possible sensitivity to this assumption was tested by assuming a ten-fold increase in dissolution rate of the fuel matrix (i.e., a one-tenth lifetime) and it was shown that the time and magnitude of peak releases to the biosphere were essentially unchanged because the reference waste-form lifetime of 50,000 years was already significantly shorter than the transport time through the geosphere [Ref. 9, p. 325]. Repository performance showed a somewhat greater sensitivity to increases in the degradation rate (decreased lifetime) of high-level glass waste because its reference lifetime for the analysis was longer, “on the order of a few hundred thousand years” [Ref. 9, p. 222]. The reference lifetime was based on the degradation rate slowing to a residual rate when the surrounding medium becomes saturated with silica. The sensitivity analysis applied an alternative conceptual model in which glass degradation rates were held constant in time at the initial rate, thus diminishing the waste-form lifetime to thousands of years. Results for this alternative model showed an insignificant increase in the peak biosphere release of the dominant radionuclide contributing to dose, I-129, from 8.6×10^{-4} mol/yr to 9.1×10^{-4} mol/yr, but a substantial shift in the time of peak release from 460,000 yr to 250,000 yr [Ref. 9, table 5.5-24].

Results from a preliminary safety assessment for the Swedish granite repository proposed at Forsmark[28] show that, for this example, transport from the repository to the biosphere can occur by relatively rapid advective flow in fractures (on the order of thousands of years). In the base case analyses for corrosion failure of waste packages [Ref. 28, Section 10.6.5], spent fuel fractional dissolution rates in the reducing environment ranged from 10^{-6}/yr to 10^{-8}/yr, corresponding roughly to waste-form lifetimes ranging from 1,000,000 yr to 100,000,000 yr [Ref. 29, Section 2.5.5]. The sensitivity analyses of the fuel dissolution rate [Ref. 28, Figure 10-44] for cases where waste package failure occurs at 500,000 years, indicate that, within the above range of values anticipated for reducing conditions, estimated dose to an exposed individual in the biosphere varies essentially linearly with the dissolution rate. This sensitivity to longer waste form lifetimes relative to the transport times is consistent with the sensitivity shown in the French repository above for longer glass lifetime, but is more pronounced in the Forsmark example because the waste form lifetime is orders of magnitude longer than the transport time. However, for the Forsmark sensitivity analyses using significantly higher dissolution rates (i.e., 10^{-5}/yr and higher – waste form lifetimes roughly 100,000 yr and less), the results are insensitive to waste form lifetime even though the transport time is much shorter. This is because the radionuclide dominantly contributing to the estimated dose, Ra-226, is continuously produced in the waste form from decay of Th-230 (which in turn is produced by decay of U-234). The rate of Ra-226 ingrowth presumably determines the availability of Ra-226 for transport at higher fuel dissolution rates, causing the overall dose to be insensitive to increases in fuel dissolution rate above 10^{-5}/yr.

Other disposal concepts show behavior comparable to that explained in detail for French and Swedish concepts. In summary, waste form durability becomes an important contributor to overall repository performance for disposal concepts where transport time to the biosphere can be relatively short compared to the regulatory period. In concepts where transport from the repository to the biosphere is dominated by slow diffusion that occurs over durations that are substantial fractions of the regulatory period, as in argillite, salt, unfractured crystalline rocks, and deep boreholes, changes in the degradation rate of the waste form may have relatively little impact on the magnitude of the estimated peak dose.

4. CONCLUSION

Insights from published safety assessments for disposal of spent nuclear fuel and high-level radioactive waste suggest that modifications to waste forms from potential advanced fuel cycles are not essential for demonstrating safe long-term performance of repositories. Modifications that reduce the thermal power of the waste or that reduce waste volume without increasing thermal loading have potential to allow more efficient use of underground mined repository galleries, and potentially also offer pathways to developing waste forms that would fit within deep borehole disposal systems. Changes in the radionuclide inventory of waste forms from the potential recovery and reuse of fissile material contained in spent fuel are unlikely to have a significant impact on the estimates of long-term performance for most disposal concepts (in the absence of disruptions that expose the waste directly to the biosphere such as human intrusion) because of the relatively higher mobility of the long-lived fission product I-129 in most disposal system environments. Waste form modifications for durability have the potential to improve estimated peak dose performance of repositories only if the modified waste-form lifetime becomes relatively long compared to the geosphere transport time, and/or approaches the period of performance (e.g., on the order of hundreds of thousands of years). Relatively smaller improvements in waste-form lifetime (e.g., on the order of thousands or tens of thousands of years) may simply delay the time of the estimated peak release to the biosphere.

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