

Summary of the Potential Impacts of Nuclear Fuel Cycle Options on the Permanent Disposal of Spent Fuel in the U.S.

S. David Sevougian, Peter N. Swift, David C. Sassani*
Sandia National Laboratories, Albuquerque, NM USA 87185
*Email (and corresponding author): dsassan@sandia.gov

All options for generating power from nuclear energy produce radioactive waste products that will require permanent isolation from the accessible environment. 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 waste characteristics such as mass, volume, radioactivity, and thermal power. Reprocessing can reduce the actinide content, volume, and thermal power of the final waste product, while various treatments of the remaining waste can create more durable waste forms. However, no options eliminate the need for robust isolation of the radioactive waste forms, which by longstanding international consensus is to be achieved by deep geologic disposal. Safety assessments for geologic disposal concepts proposed in the United States, Sweden, France, Canada, and other nations provide insight into potential benefits from reprocessing on the long-term, post-closure performance of geologic repository systems. These waste disposal concepts include mined geologic repositories in multiple host-rock types (e.g., argillite, crystalline, salt) in both saturated and unsaturated environments.

In the absence of disruptions that directly expose waste to the biosphere (such as human intrusion), estimates of long-term repository performance are dominated by radionuclides that are most mobile in groundwater, rather than those that contribute the most to total radioactivity. 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 aqueous environments, and published safety assessments for most such disposal concepts show essentially zero direct contribution to risk (in terms of estimated dose) from Pu, U, and other radionuclides proposed to be removed by various reprocessing methods. Risk in most saturated geologic disposal 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.

More specifically, radionuclide inventory reductions due to recovery and reuse of fissile isotopes of Pu and U can reasonably be expected to have little or no effect on estimates of the long-term performance of repositories in argillite^{1,2} or salt,^{3,4} or in crystalline rock concepts (mined⁵ or drilled⁶) in which transmissive fractures do not directly intersect the waste emplacement region. In each case, reducing chemical conditions immobilize actinides and long groundwater transport times prevent all but long-lived mobile species, dominated by I-129, from reaching the biosphere in significant quantities. For mined repositories in fractured granitic or crystalline rock, where relatively rapid transport to the biosphere may occur, removing the actinides that decay to create comparatively 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.⁷ Similarly, removing all actinides from the inventory of a repository in oxidizing conditions could decrease the estimated total dose by perhaps a factor of ten before I-129 becomes the primary contributor.⁸ It should be noted, however, that the currently estimated doses for concepts in either fractured rock and/or oxidizing groundwater, which include fissile actinides, are well below regulatory limits such that there is no reason to suggest they would need to be reduced further to meet safety requirements.

Waste volume and thermal power can be considered 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 (e.g., related to

the longevity of waste package and buffer materials). 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 existing repository design approaches to keep peak post-closure repository temperatures below a specified value, which do not require 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.^{9,10} Thus, thermal design constraints do not appear to limit implementation of any of the major disposal concepts for waste forms over a broad range of thermal output. 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.

Impacts of waste form durability may be evaluated for disposal of various forms of untreated spent fuel, e.g., conventional light-water reactor (LWR) uranium oxide pellets¹¹ and TRISO particles,¹² as well as vitrified waste arising 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,¹² but because many other factors (e.g., water flux through engineered barriers; degradation rates of the engineered barriers; water chemistry contacting engineered barriers and the waste form; and radionuclide transport properties) affect the timing and magnitude of the radionuclide source-term and subsequent radionuclide migration to the biosphere, the impacts of waste-form durability (i.e., lifetime) must be evaluated in the context of the full disposal system. For example, increasing the durability of a waste form may have little impact on the magnitude of the estimated peak dose if the waste form lifetime is relatively short compared to the transport time to the biosphere. This occurs in repository concepts where transport from the repository to the biosphere is dominated by slow diffusion that occurs over periods that are substantial fractions of the regulatory period, as in argillite, salt, unfractured crystalline rocks, and deep boreholes.

The potential environmental benefits of reprocessing in the U.S. are constrained by the large legacy inventory of commercial SNF from the many-decade, LWR, open fuel cycle. In particular, there are approximately 80,000 MTHM (metric tons heavy metal) of commercial SNF in storage in the U.S. as of December 2017, with about 2200 MTHM of new SNF generated nationwide each year. To date there has not been a viable economic model in which this legacy spent fuel gets reprocessed in the U.S. For example, the U.S. currently has no commercial reprocessing capability. To handle the new yearly inventory of SNF generated in the U.S. (2200 MTHM) would require two of the largest reprocessing facilities ever built, such as either the one in Sellafield, UK or the one in La Hague, France, each of which has a capacity of about 1100 MTHM/year. Thus, it has been concluded that 98% of the total current U.S. inventory by mass can proceed to permanent disposal without the need to ensure retrievability for reuse or research;¹³ and that assessment does not assume any decision about future fuel cycle options or preclude any potential options, including those with potential recycling of commercial SNF, since the 2200 MTHM that is generated annually could provide the feedstock needed for deployment of alternative fuel cycles.

In conclusion, 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 geologic 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.¹⁴ 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, or if it approaches the regulatory period of performance. For all proposed geologic disposal concepts the potential benefits of alternative fuel cycle choices should be considered in the context of additional decision criteria (i.e., in addition to waste management), including capital and operational costs, public/stakeholder acceptance, resource utilization, non-proliferation, and energy system diversification.

This summary paper is distilled in large part from work previously reported in Ref. 14. Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA-0003525. The views expressed in this paper do not necessarily represent the views of the U.S. Department of Energy or the United States Government.

1. NAGRA (Nationale Genossenschaft für die Lagerung Radioactiver Abfälle) 2002. *Project Opalinus Clay Safety Report: Demonstration of disposal feasibility for spent fuel, vitrified high-level waste and long-lived intermediate-level waste (Entsorgungsnachweis)*, Technical Report 02-05.
2. ANDRA (French National Radioactive Waste Management Agency) 2016. *Safety Options Report – Post-closure Part (DOS-AF)*, CG-TE-D-NTE-AMOA-SR2-0000-15-0062/A, July 11, 2016, 466 pp., https://international.andra.fr/sites/international/files/2019-03/Safety%20Options%20Report%20-%20Post-Closure_1.pdf
3. DOE (U.S. Department of Energy) 2014. *Title 40 CFR Part 191 Subparts B and C Compliance Recertification Application 2014 for the Waste Isolation Pilot Plant*, DOE/CA) 14-3503.
4. von Berlepsch, T., and B. Haverkamp 2016. "Salt as a Host Rock for the Geological Repository for Nuclear Waste, *Elements*, v. 12, p. 257-262.
5. NWMO (Nuclear Waste Management Organization) 2013. *Adaptive Phased Management: Postclosure Safety Assessment of a Used Fuel Repository in Sedimentary Rock*, NWMO TR-2013-07, Figure 7-96, Toronto, Canada.
6. Freeze, G., E. Stein, L. Price, R. MacKinnon, and J. Tillman 2016. *Deep Borehole Disposal Safety Analysis*. FCRD- UFD-2016-000075 Rev. 0; SAND2016-10949R. U.S. Department of Energy, Washington, DC.
7. SKB (Svensk Kärnbränslehantering AB) 2011. *Long-term Safety for the Final Repository for Spent Nuclear Fuel at Forsmark: Main Report of the SR-Site Project*, Technical Report TR-11-0, Figure 13-18.
8. DOE (U.S. Department of Energy) 2008. *Yucca Mountain Repository License Application*, DOE/RW- 0573, Rev. 1, Fig. 2.4-20b.
9. Hardin, E. L. and E. Kalinina 2016. *Cost Estimation Inputs for Spent Nuclear Fuel Geologic Disposal Concepts (Revision 1)*. SAND2016-0235. Sandia National Laboratories, Albuquerque, NM.
10. Hardin, E. L., T. Hadgu, D. Clayton, R. Howard, H. Greenberg, J. Blink, M. Sharma, M. Sutton, J. Carter, M. Dupont, and P. Rodwell 2012. *Repository Reference Disposal Concepts and Thermal Load Management Analysis*. FCRD-UFD-2012-000219 Rev. 2. Sandia National Laboratories, Albuquerque, NM.
11. Sassani, D. and F. Gelbard 2019. "Performance assessment model for degradation of tristructural-isotropic (TRISO) coated particle spent fuel," *Proceedings of the American Nuclear Society International High-Level Radioactive Waste Management Conference*, April 14-18, 2019.
12. Peters, M. T. and R.C. Ewing 2007. "A science-based approach to understanding waste form durability in open and closed nuclear fuel cycles," *Journal of Nuclear Materials* v. 362, p. 395-401
13. Wagner, J. C., J.L. Peterson, D.E. Mueller, J.C. Gehin, A. Worrall, T. Taiwo, M. Nutt, M.A. Williamson, M. Todosow, R. Wigeland, W. Halsey, R. Omberg, P.N. Swift, and J.T. Carter, 2012. *Categorization of Used Nuclear Fuel Inventory in Support of a Comprehensive National Nuclear Fuel Cycle Strategy*, ORNL/TM-2012/308, FCRD-FCT-2012-000232, Oak Ridge National Laboratory, Oak Ridge, TN, USA, December 2012.
14. Swift, P. N. and D. C. Sassani 2019. "Impacts of Nuclear Fuel Cycle Choices on Permanent Disposal of High-Activity Radioactive Wastes," in *Proceedings of the IAEA International Conference on the Management of Spent Fuel from Nuclear Power Reactors*, 24-28 June 2019, Vienna, Austria, IAEA-CN-272-185, SAND2019-5941C.