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PERFORMANCE ASSESSMENT ANALYSES UNIQUE TO DEPARTMENT OF ENERGY SPENT NUCLEAR FUEL

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ABSTRACT

This paper describes the iterative process of grouping and performance assessment that has led to the current grouping of the U.S. Department of Energy (DOE) spent nuclear fuel (SNF). The unique sensitivity analyses that form the basis for incorporating DOE fuel into the total system performance assessment (TSPA) base case model are described. In addition, the chemistry that results from dissolution of DOE fuel and high level waste (HLW) glass in a failed co-disposal package, and the effects of disposal of selected DOE SNF in high integrity cans are presented.

I. INTRODUCTION

The potential Monitored Geologic Repository at Yucca Mountain contains 70,000 metric tons heavy metal (MTHM) of high-level radioactive wastes composed of 7,000 MTHM of defense waste and 63,000 MTHM of commercial SNF. The defense waste consists of 2.333 MTHM of DOE SNF and 4,667 MTHM of HLW. The DOE SNF is composed of more than 250 different fuel types with a wide range of characteristics such as fuel matrix, fuel configuration, fissile content, burnup, cladding, and fuel and cladding integrity. In the mid-1990s the National Spent Nuclear Fuel Program began an iterative process of assessment and re-evaluation that has progressed to the performance assessment calculations that will be conducted to incorporate the DOE SNF into the Site Recommendation (SR) and License Application (LA). These analyses are unique due to the unique character of the fuels themselves, the unique co-disposal approach for DOE SNF and HLW, and the use of surrogate fuels to represent the DOE spent nuclear fuel (SNF) in the TSPA base case model.

II. SPENT NUCLEAR FUEL GROUPS

The factors used to group the different types of DOE SNF have different levels of importance depending on the specific analysis that will be preformed based on the grouping results. The factors used in grouping are fuel matrix, fuel cladding, fuel condition, fissile species enrichment, and burnup. Each factor has a different level of importance depending on whether the grouping is being performed for design basis accident analysis, criticality analysis, or performance assessment. For example, fissile content and species is more important to grouping for criticality than it is to grouping for performance assessment. Application of the factors important to performance assessment yields the grouping that is used for the assessment.

The iterative process of grouping followed by performance assessment has led to 11 groups¹ of DOE SNF as follows where the number in parenthesis is the total amount of SNF in each group in MTHM:

- 1. Classified Navy (65)
- 2. Plutonium/Uranium alloy (9.1)
- 3. Plutonium/Uranium carbide (0.1)
- 4. Plutonium/Uranium & Plutonium oxide (12.4)
- 5. Thorium/Uranium carbide (26.3)
- 6. Thorium/Uranium oxide (50.4)
- 7. Uranium metal (2,127.2)
- 8. Uranium oxide (178.2)
- 9. Aluminum based (20.9)
- 10. Unknown (4.5)
- 11. Uranium-Zirconium hydride (1.6).

This grouping is somewhat different than the grouping used for the total system performance assessment for the viability assessment (TSPA-VA) that contained 16 SNF groups.^{2, 3} It is also somewhat different than that of an earlier set of analyses conducted prior to TSPA-VA that analyzed 13 groups of SNF.⁴ The groupings have been simplified through the iterative process of grouping and performance assessment. The results of the performance assessment have been used to simplify the next grouping. The iterative process has shown that the only grouping parameter important to performance is fuel matrix. However, burnup is used to obtain the radionuclide inventory for each SNF group. Even though the overall groupings have been simplified the major results of each successive performance assessment are unchanged.^{4, 3, & 5}

In the past, the total amount (2,495.7 MTHM) of SNF has been reduced (with the exception of Navy fuel) to the 2,333 MTHM incorporated into the base case TSPA model. However, sensitivity analyses have been conducted for the full amount of DOE SNF and the HLW necessary to co-dispose that amount of SNF.^{4, 3, & 5} The grouping shown above was used for the latest sensitivity analyses of DOE SNF,⁵ and will be used for the analyses supporting the Site Recommendation (SR).

III. PERFORMANCE ASSESSMENT

Performance assessment is conducted using a simplified model that incorporates all of the processes and events important to safety. This model incorporates the potential wastes disposed at the repository, the repository design, the waste package design, the geologic and hydrologic systems, and the biosphere. The Repository Integration Program (RIP) model captures uncertainty of the processes and events through statistical distributions of model parameters. Analyses are conducted using the probabilistic model to analyze doses from numerous sets of natural and engineered system conditions that in total represent the potential behavior of the overall system. The complex nature of the model limits the number of waste forms explicitly incorporated into it due to restricted computer storage and model running time. For these reasons the DOE SNF is incorporated into the base case TSPA model as a surrogate that is representative of the DOE SNF groups.

The base case TSPA model was used to conduct sensitivity analyses of the individual groups of DOE SNF to show that the surrogate used is a reasonable representation. Sensitivity analyses were conducted for each group, one at a time, in the environment of the base case model by calculating the expected value dose history attributed to that group. The dose history for all DOE groups was then summed and compared to the dose history for the surrogate used in the base case model. Sensitivity analyses of physical properties, dissolution models, and dissolution rates are used to demonstrate that

the dissolution model used for the surrogate is conservative as compared to more realistic representations. For TSPA-VA the DOE SNF, including Navy fuel, was incorporated in the base case model as a single uranium metal surrogate, and the uranium metal dissolution model was used for the surrogate fuel.³ The sensitivity analyses used dissolution models for specific groups (carbide, oxide, ceramic, or uranium metal).³ The dose from each group was summed and the composite of all groups was compared to the dose from the surrogate used in the base case model to indicate that the surrogate was representative of the DOE fuels. The composite of the individual groups was also compared to an equivalent amount of unclad commercial SNF.³ In the 1999 analyses of DOE SNF the grouping above was used to investigate disposal of small amounts of degraded SNF in a high integrity can (HIC); the effects of including radionuclides in addition to the nine analyzed in TSPA-VA; and the geochemical effects of HLW and SNF dissolving in a failed co-disposal waste package.

The fissile content of much of the DOE fuel will necessitate co-disposal with five canisters of HLW glass surrounding one canister of SNF. Co-disposal is unique to DOE fuel, and the dissolution of fuel and glass combined with corrosion of metal package components can change the radionuclide release rate from the failed package. In the 1999 performance assessment, the geochemical model EQ3/6 was used to analyze the effects of co-disposal on release of radionuclides from the failed package.⁵

IV. RESULTS

The expected-value dose history of an individual 20 km down the hydraulic gradient from the repository is illustrated in Figure 1 for the TSPA-VA base case model. The initial dose, from first occurrence to approximate 40,000 yr is from a combination of 99 Tc and 129 I, and after 50,000 yr the dose is primarily from 237 Np (Figure 1).

Figure 2 depicts a comparison of the expected value total dose histories at 20 km over 100,000 yr from 2,333 MTHM of each of the following: the surrogate uranium metal fuel used in the base case model; the total dose from the composite of the individual SNF groups; and unclad commercial SNF.³ These results suggest that the uranium metal surrogate is a good representation of the DOE fuel in the base case model, and that unclad commercial SNF could be used as an upper bound for the DOE SNF. The ragged nature of the composite dose curve is due to the method of development. Spikes in dose, caused by rapid release of ⁹⁹Tc and ¹²⁹I, at failure of individual packages are additive for groups having similar numbers and twice the number of packages. The package



Figure 1. Expected-value dose history at 20 km over 100,000 yr from TSPA-VA base case.



Figure 2. Expected-value dose history at 20 km over 100,000 yr from 2,333 MTHM of DOE SNF, from 2,333 MTHM of the Surrogate DOE SNF used in the Base Case, and from 2,333 MTHM of Commercial SNF.

failures, at approximately the same time, produces spikes in the composite curve that may be unrealistically high. The results of TSPA-VA³ also indicated that the dose from Navy fuel was about two orders of magnitude below that from an equivalent dose (65 MTHM) of unclad commercial SNF. Due to the robust nature of the Navy fuel, its dose contribution may be very conservatively bounded by the unclad uranium metal surrogate or unclad commercial SNF.

Analyses of radionuclides in addition to those included in TSPA-VA were conducted by calculating the dose at the water table below the repository. This calculation was done because there are no breakthrough curves in the model for transport of the added radionuclides through the saturated zone. Where breakthrough curves exist for radionuclides, comparison of dose at the water table and dose 20 km from the repository shows that transport through the saturated zone reduces dose by approximately a factor of ten. Analyses of thorium/uranium carbide SNF (Group 5, where Ft. St. Vrain fuel is typical) indicates that 229 Th and 233 U contribute significantly to the total dose (at the water table) after about 250,000 yr. In addition, after 250,000 yr these radionuclides produce a larger contribution to total dose than that from ²³⁷Np. Based on these results the actinides in the ²²⁹Th chain are being incorporated into the TSPA-SR base case model.

Sensitivity analyses of dissolution rate show that there is no difference in dose history when the uranium metal dissolution model is used or instantaneous dissolution is assumed. This lack of difference exists because the ⁹⁹Tc and ¹²⁹I in the waste package are released in less than one model time step for both cases, and the release of ²³⁷Np is controlled by solubility irrespective of how much is available for transport. This result would also be obtained for the oxide dissolution model for commercial SNF as long as no cladding is assumed. The assumption of cladding would moderate the release by the cladding failure rate. Due to the amount of disrupted cladding on most of the DOE SNF, no credit is taken for cladding for the surrogate SNF. Sensitivity analyses of percent of cladding present for DOE SNF indicate that this is a conservative assumption. For example, 50 percent cladding present on Group 7 (N Reactor) SNF would reduce the height of the spikes caused by ⁹⁹Tc and ¹²⁹I by a factor of two, but does not change the dose from ²³⁷Np because there is more available for transport than can be transported at its solubility limit.

Regarding the analyses of the HIC, an Alloy 22 can with a wall thickness of 0.25 in.^5 was assumed. Figure 3 depicts the results of disposal of one HIC per co-disposal package compared with not using the HIC. This figure also depicts the dose from the TSPA-VA base case model for the entire repository. The added corrosion resistant barrier when the HIC is used delays the peak dose by about 60,000 yr (Figure 3). The later dose peak has the same dose rate as the earlier peak due to the long half-life of ²³⁷Np (2.14 x 10⁶ yr). When the HIC is not used the fuel is assumed to be disposed in a stainless-steel canister that provides no time delay of release after the waste package has failed (i.e., the failure of the canister would occur in less than one model time step).

Analyses of the geochemical environment in a failed co-disposal package indicate that the geochemistry, shortly after failure, is dominated by the corrosion of A516 carbon steel (for about 300 yr for Fast Flux Test Facility fuel) greatly reducing the pH (Figure 4).⁵ The corrosion of the steel components of the waste package is tempered by the alkalinity resulting from the dissolution of the HLW glass. These two effects cause the geochemistry within the failed package to be very sensitive to the assumed corrosion and dissolution rates.

V. CONCLUSIONS

Results of analyses of DOE SNF using the TSPA-VA base case model and the geochemical model EQ3/6 are as follows:

- Grouping of DOE SNF for performance assessment depends only on fuel matrix. However, fuel burnup is used to obtain the radionuclide inventories needed for the performance assessment model.
- Sensitivity analyses of DOE SNF demonstrate that Groups 2-11 can be represented by an unclad uranium metal surrogate in the TSPA base case model, and that unclad commercial SNF surrogate would bound the behavior of these groups. Due to the robust nature of Navy fuel, clad commercial SNF may be more a appropriate surrogate for Group 1 SNF and it will be used in the TSPA-SR base case model.⁶
- Sensitivity analyses indicate that there is no difference in dose when assuming instantaneous fuel dissolution or the uranium metal dissolution model.



Figure 3. Comparison of the TSPA-VA base case over 1,000,000 yr with the dose using the high integrity can and the dose not using the high integrity can.



Figure 4. Radionuclide aqueous concentrations, waste package materials remaining, and pH versus time from a waste package breach for the Fast Flux Test Facility SNF (Group 4).

- Examination of adding the actinides in the ²²⁹Th chain to those radionuclides considered in TSPA-VA shows that for Group 5 SNF ²²⁹Th and ²³³U are significant contributors to dose after about 250,000 yr. As a result of this finding, actinides in this chain will be added to the radionuclides considered for TSPA-SR.
- Use of an Alloy 22 high integrity can for disposal of SNF delays the dose peak by about 60,000 yr.
- Analyses of geochemistry in the failed co-disposal waste package show that the corrosion of steel has a significant influence on pH and radionuclide release. This corrosion effect is moderated by the alkalinity of the HLW glass.

Results using the TSPA-SR base case model are expected to be similar to those of earlier TSPA models. However, the dose peaks will be moved farther out in time due to the addition of a drip shield that increases the waste package lifetime.⁷ The conclusions drawn using the TSPA-SR base case model are not expected to change as the dose curves would have the same relationship with each other though dose peaks occur at later times.

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