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Repository Performance Assessment of Waste Forms from the Electrometallurgical  
Treatment of Sodium-Bonded Spent Nuclear Fuel\*

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REPOSITORY PERFORMANCE ASSESSMENT OF WASTE FORMS FROM THE  
ELECTROMETALLURGICAL TREATMENT OF SODIUM-BONDED SPENT NUCLEAR FUEL

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## ABSTRACT

The ceramic and metal waste forms produced by electrometallurgical treatment of sodium-bonded spent nuclear fuel are undergoing evaluation as to how they will perform within the geologic repository which is proposed to be built at Yucca Mountain. An initial assessment, making use of preliminary degradation models for the waste forms, is described. The analyses are performed with a simplified version of the Total System Performance Assessment - Viability Assessment repository model. Results indicate that the ability of the ceramic and metal waste forms to retain radionuclides is similar to and sometimes better than defense high-level waste glass.

## I. INTRODUCTION

An electrometallurgical treatment for spent nuclear fuel is under development at Argonne National Laboratory (ANL). The work described here was carried out to provide an initial assessment of the repository performance of the waste that results when the treatment is applied to sodium-bonded, DOE spent nuclear fuel containing 60 metric tons of heavy metal (MTHM). It also provides an indication of the adequacy of the data which will be supplied by the Laboratory in response to DOE data calls for the Yucca Mountain Project (YMP) Site Recommendation and the License Application. When applied to sodium-bonded spent nuclear fuel, the electrometallurgical treatment produces ceramic and metal waste forms. Both waste forms are planned for ultimate disposal in the proposed Yucca Mountain repository.

In the electrorefining process, molten LiCl-KCl salt is used to accumulate sodium, fission products, and transuranics from the sodium bonded spent fuel. Periodically, the salt must be discarded or recycled. Discarded salt is immobilized in a ceramic waste form consisting of glass bonded sodalite encased in steel cans.

The cans are called "HIP cans" because they and their contents have been subjected to hot isostatic pressing to consolidate the waste form.

The metal waste form consists of ingots containing primarily stainless steel cladding hulls, zirconium, and noble metal fission products. The ingots also contain some transuranics and other fission products, including  $^{99}\text{Tc}$ .<sup>1</sup>

All calculations described here were carried out using the Repository Integration Program (RIP).<sup>2</sup> RIP was used by the YMP to carry out the Total System Performance Assessment - Viability Assessment (TSPA-VA).<sup>3</sup> RIP is a probabilistic simulator for modeling environmental systems. The computer program allows input parameters to be either deterministic or stochastic. It can be run in an expected-value mode in which all stochastic input parameters are set to their expected values or in a Monte Carlo mode in which calculations are performed for several independently sampled sets of values for the stochastic input parameters. A key parameter calculated by the RIP viability assessment model was the estimated dose received by an individual using water from a well 20 km from the repository. However, the RIP code was also used to estimate the release of radionuclides across various boundaries within the mountain.

A simplified version of the YMP performance assessment model was used for the current analysis. Calculated dose rates using this model are in very good agreement with the more detailed viability assessment model and results are obtained as much as ten times faster. Preliminary degradation models for the ceramic and metal waste forms were added to the simplified model for this work. Preliminary degradation models for the waste forms were used because of the limited experimental degradation results available at the time this initial assessment was performed. As more detailed experimental information becomes available and the degradation models are revised,

it will be necessary to repeat some of the analyses described here.

The current assessment indicates that the ANL waste forms will retain radionuclides as well as or better than the borosilicate glass used for defense high-level waste (DHLW) as modeled in the viability assessment. They generally do not perform as well as the cladded commercial spent nuclear fuel as modeled in the viability assessment. ANL waste forms will make a negligible contribution to the dose rate at the 20-km well considered in the viability assessment.

## II. THE SIMPLIFIED MODEL

At the request of ANL, Golder Associates, Inc. (GAI) developed a simplified version of the performance assessment model constructed by the YMP for the viability assessment.<sup>4</sup> The simplified version retained the model used in the viability assessment for the engineered barrier system (the waste package and structural material within the tunnel). However, it describes transport through the unsaturated and saturated zones using models that are built into RIP. In the viability assessment, transport through the unsaturated zone is described with the FEHM particle tracking model.<sup>5</sup> Transport through the saturated zone is described using a convolution model.<sup>6</sup>

In documenting the simplified model, GAI demonstrated that the model faithfully reproduced the results obtained with the more detailed model used by the YMP. Figure 1 shows a comparison of the dose rate at the 20-km well between the simplified model and the YMP model for a 1,000,000-year simulation. For the case shown, all stochastic input variables were set at their expected values. The results shown indicate very close agreement except for the peaks associated with climate changes postulated by the viability assessment to occur approximately every 100,000 years.

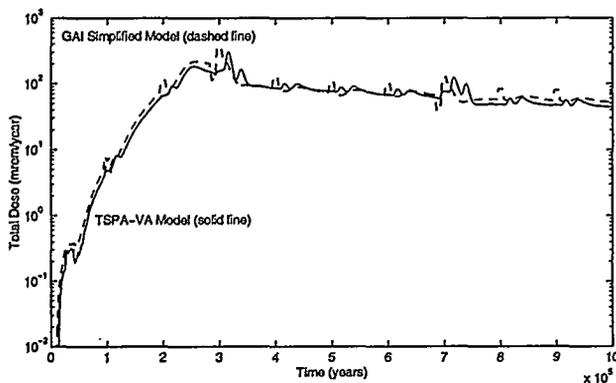


Fig. 1. Comparison of dose rates at the 20-km well calculated with the GAI simplified model and with the YMP TSPA-VA model.

For the analysis describe here, two additional source terms, one each for the ceramic and metal waste forms, were added to the simplified model provided by GAI. The TSPA-VA analysis used nine radionuclides to represent the commercial spent nuclear fuel, high-level defense glass, and DOE spent nuclear fuel waste forms. In contrast, the earlier 1995 total system performance assessment (TSPA-95) analysis performed by the YMP made use of 39 radionuclides to represent the waste forms.<sup>7</sup> In adding source terms to represent the ANL waste forms, screening calculations were first performed to verify that the waste forms could be adequately characterized by the 39 radionuclides used in the TSPA-95 analysis. Then 39- and nine-isotope simplified models were used to demonstrate that the wastes forms could also be adequately characterized using the nine radionuclides selected by the YMP for the TSPA-VA analysis.

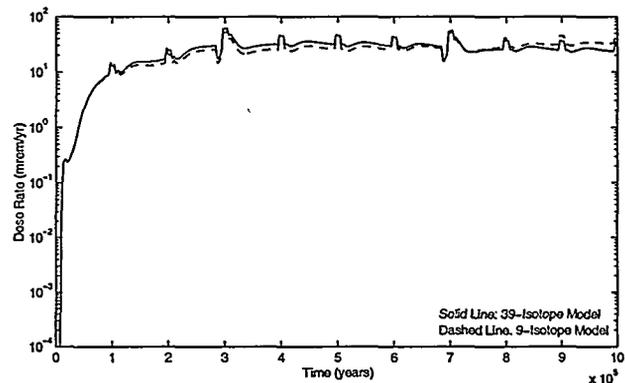


Fig. 2. Comparison of dose rates calculated with 39- and nine-isotope models containing only ANL waste forms.

Figure 2 compares the dose-rate time history at the 20-km well calculated with a 39-isotope simplified model with the corresponding time history calculated with a nine-isotope simplified model. Expected values were used for all stochastic input parameters. For the comparison, the simplified models contained only ANL waste forms in each region of the repository. The number of waste packages was arbitrarily increased so that the total mass of <sup>99</sup>Tc was approximately the same as the mass of <sup>99</sup>Tc in the commercial spent nuclear fuel included in the TSPA-VA model. This was done so that the ANL waste forms had fission products from approximately the same number of fissions as had occurred in the commercial spent nuclear fuel. Both ANL waste forms were assumed to degrade at the same rate as the DHLW glass in the TSPA-VA model. The comparison in Fig. 2 indicates that the dose rate contribution from the ANL waste forms can be adequately approximated using the nine-isotope model. The nine-isotope model underpredicts the dose rate in the time period from about 100,000 to 700,000 years because several radionuclides, each of which contribute on the order of 1% of the dose, are not included in the model. It overpredicts

the dose rate after about 750,000 years because the model provides a poor representation of the dose-rate contribution from  $^{231}\text{Pa}$ . Additional information regarding the 39-isotope model and comparisons with the nine-isotope model can be found in a more detailed report.<sup>8</sup>

### III. WASTE FORM DEGRADATION MODELS

The calculations discussed in the remainder of this paper make use of preliminary waste-form degradation models developed specifically for the ANL waste forms. The RIP computer program requires a matrix degradation rate which defines the fraction of the undegraded waste form which degrades per unit time. The analysis here follows the pattern established in the TSPA-VA and evaluates the matrix degradation rate as the product of the specific area of the waste form and a rate describing the mass that degrades per unit surface area per unit time. The specific area of the ceramic waste form is defined as the surface area of the ceramic in a single HIP can divided by the mass of ceramic in the can. Cracking in the waste form is accounted for by multiplying the specific area by a cracking factor of five. This factor is based on measurements for HIP cans smaller than those destined for the repository. DHLW glass was assumed to have a cracking factor of 21 in the TSPA-VA. The specific area of the metal waste form is the surface area of a single metal ingot divided by the mass of the ingot. The specific areas are assumed to remain constant as the waste forms degrade. This assumption was also made for the waste forms analyzed in the TSPA-VA.

The rate expression used for the ceramic waste form is the same as the rate expression used to represent DHLW glass in the TSPA-VA model. For the analyses presented here, rate data used in the TSPA-VA is used to represent the ceramic waste form. Dissolution rates are calculated based on incoming groundwater composition and waste-package temperatures. In-package chemistry changes are not represented. This is consistent with the TSPA-VA model for DHLW glass. The model does, however, use the specific surface area and cracking factor as determined for the ceramic waste form. As described in a companion paper,<sup>9</sup> experimental work to measure the dissolution rates of the ceramic waste form is in progress. Current results suggest the dissolution rate of the ceramic waste form will be similar to that of the other high-level waste glasses.

Like the DHLW glass model, the ceramic waste-form model does not take credit for the retention of radionuclides in alteration phases. As a result, radionuclides are made available for transport once released from the waste-form matrix. In addition, the ceramic waste-form model takes no credit for the durability of the HIP can.

Release rates from the metal waste form will depend on three interrelated factors: the metallurgy of the metal

waste form, the degradation mechanism, and the environmental conditions to which the metal waste form is exposed. The environmental conditions—and, thus, the degradation mechanisms—are likely to change over time in the repository. Current experimental studies indicate similar corrosion behaviors for the metal waste form and type 316 stainless steel. Therefore, the metal waste-form degradation modeling has presumed that the degradation *mechanisms* are the same as those for stainless steel. However, the degradation *rates* may differ.

Eleven potential corrosion mechanisms were identified for stainless steel under repository conditions. Of these mechanisms, two are expected to dominate the release of radioactive isotopes from the metal waste form: uniform aqueous corrosion and crevice corrosion. Based on corrosion data from the literature and from ANL experiments, empirical relationships were developed to express the corrosion rates of stainless steel in terms of temperature, pH, and chloride concentration. The net corrosion rate was calculated as a linear combination of the uniform and crevice rates, weighted by the surface area fractions expected to be undergoing each corrosion mechanism. Existing corrosion data for various metal waste-form alloy compositions were compared with predictions from the stainless steel model. The empirical parameters of the model were adjusted based on the metal waste-form data. This metal waste form corrosion equation was incorporated into the simplified RIP performance assessment model for the Yucca Mountain repository. No credit was taken for the ability of corrosion products to retain radioactive isotopes. That is, it was assumed that as regions of the metal waste form degrade all radioactive isotopes originally contained in that region are immediately available for transport.

Experimental measurements in support of waste-form degradation modeling for both the ceramic and metal waste forms are ongoing. Modification of the degradation models for the ceramic and metal waste forms can be anticipated as the experimental programs progress. As these modifications occur, the analyses described in the next section will be revisited.

### IV. COMPARATIVE WASTE FORM PERFORMANCE

The TSPA-VA model divided the repository into six regions. The regions differ in area and in their thermal and moisture flow characteristics. Because of the anticipated small number of packages containing the ANL waste forms (60 MTHM contained in an estimated 25 packages of ceramic waste and 1 package of metal waste, assuming the ceramic and metal waste forms are kept in separate packages), it was arbitrarily assumed that all of the ANL waste would go into the southeast region. This is one of

the larger regions. Differences in waste form performance from region to region are expected to be small.

Because of the small amount of ANL waste, it will make a negligible contribution to the dose rate at the 20-km well.<sup>8</sup> Consequently, the waste forms were compared on the basis of the normalized cumulative release of various radionuclides from the engineered barrier system. The normalized cumulative release is defined as the cumulative release in grams (or Ci) divided by the initial inventory expressed in grams (or Ci). As noted earlier, many of the input parameters in the RIP model are stochastic. The results described in the following paragraphs are the result of averaging the cumulative releases evaluated for each of 100 independent samples of the stochastic input parameters. The averaging is carried out at each time point.

Radionuclides are introduced into the RIP calculations by means of source terms. Each waste form will be represented by one or more source terms. The source term specifies the radionuclide inventory, the number of packages, and the degradation rate for the waste form. It also specifies the failure distribution for the barriers separating the waste form from the environment. Only a single barrier is used for the ceramic, metal, high-level defense glass, and DOE spent nuclear fuel waste forms. The fraction of the waste packages that have failed at a given time is input to RIP by means of several tables. The particular table used depends on the region in the repository and on assumptions about infiltration rates. The number of packages that have failed in a given region is computed as product of the fraction of packages failed and the total number of packages. The product is truncated to the nearest integer. Two source terms in the same region use the same failure fraction. Thus, if one source contains two packages, the first package failure will occur at the time when the failure fraction is 0.5. If another source has four packages, the first package will fail at the time when the failure fraction is 0.25. Under these circumstances, the source term with the larger number of packages will always release radionuclides earlier than the source term with the smaller number of packages. To compare normalized cumulative releases from the engineered barrier system, it was decided to make the comparison for source terms in the same repository region (southeast) having the same number of packages. The number of packages was set to 1000. The large number is based on the expectation that a larger number would better represent the average behavior of the waste packages.

Figures 3 and 4 show the normalized cumulative releases for the isotopes <sup>99</sup>Tc and <sup>129</sup>I for the 10,000-year time period. These isotopes are the dominant contributors to the dose rate at the 20-km well during the first 10,000 years. In spite of the relatively large solubility of Tc, the early release of <sup>99</sup>Tc from the engineered barrier system is solubility limited. The inventory per package of <sup>99</sup>Tc in the

metal waste form is about the same as the inventory for a package of commercial spent nuclear fuel and larger than the inventories in packages of either DHLW or DOE spent nuclear fuel. For waste forms for which the release is solubility limited, the normalized cumulative release will be larger for the waste forms with the smaller inventory. Because for the ANL metal waste form the inventory per package of <sup>99</sup>Tc is comparable to that for commercial spent nuclear fuel and is larger than that for either DHLW or DOE spent nuclear fuel, the early normalized release from the metal waste form is comparable to that from cladded commercial spent nuclear fuel and much lower than the normalized releases from DOE spent nuclear fuel or DHLW. There is no initial inventory of <sup>99</sup>Tc in the ceramic waste form.

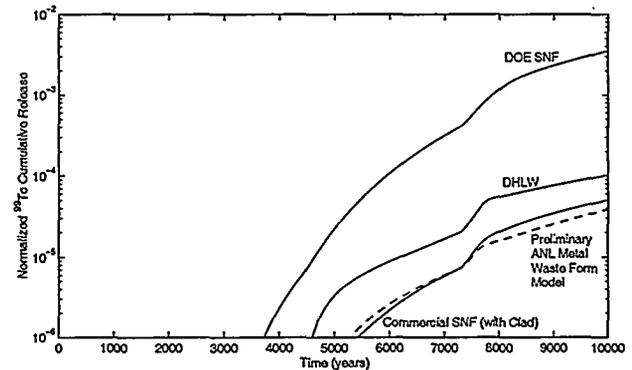


Fig. 3. Normalized cumulative release of <sup>99</sup>Tc from the engineered barrier system for various waste forms.

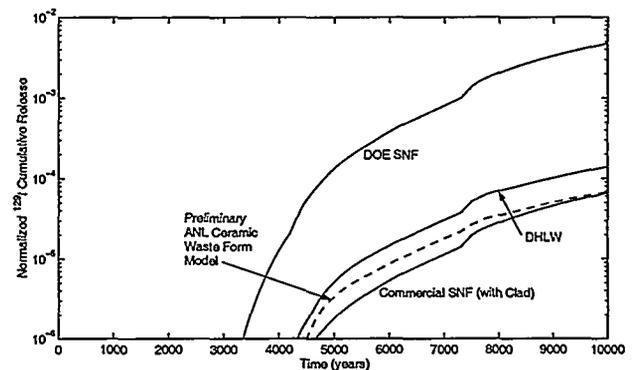


Fig. 4. Normalized cumulative release of <sup>129</sup>I from the engineered barrier system for various waste forms.

The solubility of <sup>129</sup>I is large enough so that its release from the engineered barrier is controlled by the waste-form degradation rates. Fig. 4 shows that the normalized release from the ceramic waste form is less than that from DHLW glass and, at 10,000 years is approaching the normalized release from cladded commercial spent nuclear fuel. There is no initial inventory of <sup>129</sup>I in the metal waste form. Since the rate terms for the ceramic waste-form and for DHLW glass are assumed to be the same, the difference

between the releases for the ceramic and glass waste forms can be attributed to the smaller cracking factor assumed for the ceramic waste form. The specific area is larger for the ceramic waste form than for the DHLW glass. The cracking factor used in the calculations was measured for HIP cans considerably smaller than are expected to be sent to the repository. This parameter will be reevaluated when full-scale waste forms are produced. The results shown in Figs. 3 and 4 show the importance of cladding for the performance of the commercial spent nuclear fuel. This importance is underscored in another calculation, not shown, but analogous to that in Fig. 4, in which the curve for the normalized release of  $^{129}\text{I}$  from commercial spent nuclear fuel without cladding was found to be very close to that for DOE spent nuclear fuel.

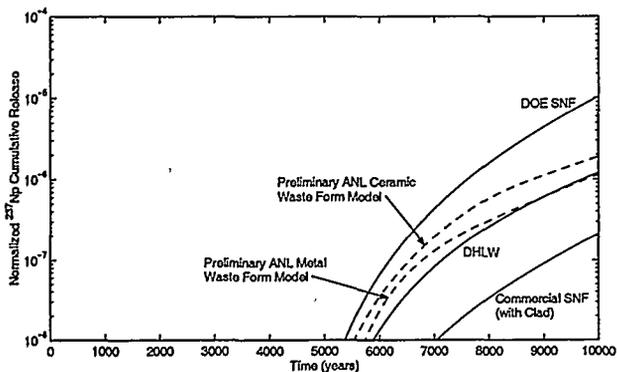


Fig. 5. Normalized cumulative release of  $^{237}\text{Np}$  from the engineered barrier system for various waste forms.

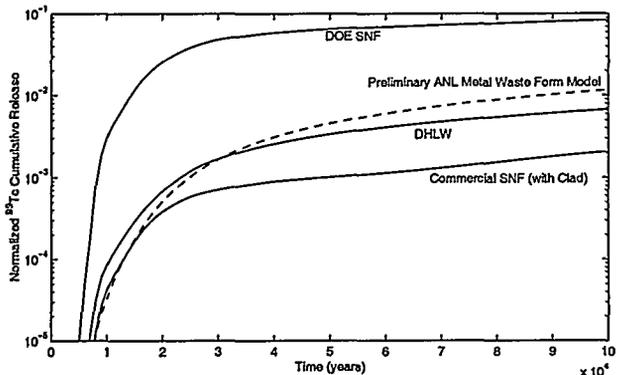


Fig. 6. Normalized cumulative release of  $^{99}\text{Tc}$  for the engineered barrier system for various waste forms.

$^{237}\text{Np}$  is present in both the metal and ceramic waste forms. Throughout the first 10,000 years, the release of  $^{237}\text{Np}$  is solubility limited for all waste forms. One consequence of this is that the normalized release of  $^{237}\text{Np}$  is lower than that for  $^{99}\text{Tc}$  or  $^{129}\text{I}$  for each of the waste forms. Another is that all the waste forms are releasing  $^{237}\text{Np}$  from the engineered barrier at the same rate. Figure

5 shows that the normalized release from the ANL metal waste form exceeds that from DHLW during most of the first 10,000 years. This occurs even though the ANL metal waste form has a lower degradation rate during most of this time period. The normalized release from the ANL ceramic waste form exceeds that from DHLW throughout the first 10,000 years. Recall that the rate parameter for the ceramic waste form dissolution is the same as that for DHLW in these calculations. The higher normalized releases are caused by the fact that the inventory of  $^{237}\text{Np}$  per package is lower in the ANL waste forms than for the other waste forms.

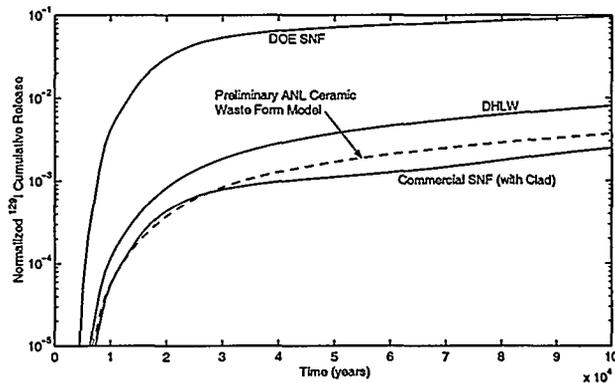


Fig. 7. Normalized cumulative release of  $^{129}\text{I}$  from the engineered barrier system for various waste forms.

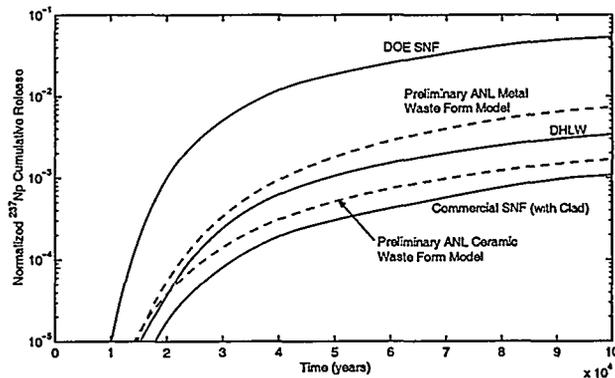


Fig. 8. Normalized cumulative release of  $^{237}\text{Np}$  from the engineered barrier system for various waste forms.

During the first 100,000 years, the contribution to the dose rate at the 20-km well passes through a transition from being dominated by the release of  $^{99}\text{Tc}$  and  $^{129}\text{I}$  to being dominated by  $^{237}\text{Np}$ . Figures 6, 7, and 8 show the normalized releases for these radionuclides over the 100,000-year time period. Figure 6 shows that the normalized release of  $^{99}\text{Tc}$  from the metal waste form is similar to the normalized release from cladded commercial spent nuclear fuel from about 8,000 years until nearly 20,000 years. It remains below the release from DHLW for

about 30,000 years and remains much lower than the release from DOE spent nuclear fuel throughout the remainder of time period. The normalized release of  $^{129}\text{I}$  from the ceramic waste form, shown in Fig. 7, is below that from DHLW glass, and from about 10,000 to 30,000 years is similar to or below the normalized release from cladded commercial spent nuclear fuel. Figure 8 shows that prior to about 20,000 years the normalized release of  $^{237}\text{Np}$  from the ANL ceramic waste form is somewhat above that from DHLW and after 20,000 years is less than that from DHLW. The figure also shows that the normalized release of  $^{237}\text{Np}$  from the ANL metal waste form is always greater than that from DHLW, but considerably less than that from DOE spent nuclear fuel. Note also that the normalized releases for  $^{237}\text{Np}$  remain lower than the releases of  $^{99}\text{Tc}$  and  $^{129}\text{I}$ , respectively, for the ANL metal and ceramic waste forms. This is consistent with its lower solubility.

Calculations have also been performed for the five waste forms over the first 1,000,000 years. After about 300,000 years, more than 70% of packages that are exposed to dripping water have failed (about 25% of packages in the southeast region of the repository are exposed to dripping water). Because of the poor performance of the DOE spent nuclear fuel matrix, essentially all of the  $^{237}\text{Np}$  inventory has been released from these failed packages by 300,000 years. In contrast, even though the same fraction of the ANL waste form packages have failed, by 300,000 years the ceramic waste form has released only about 4% of its failed-package  $^{237}\text{Np}$  inventory and the metal waste form about 30% of its failed-package  $^{237}\text{Np}$  inventory. The ceramic waste form may retain  $^{237}\text{Np}$  long enough for a significant amount of the  $^{237}\text{Np}$  to decay before reaching the 20-km well. The normalized release of  $^{99}\text{Tc}$  from the ANL metal waste form at the end of the 1,000,000-year period is significantly lower than the corresponding release of  $^{237}\text{Np}$  because of the significantly shorter half-life of  $^{99}\text{Tc}$ . The normalized release of  $^{237}\text{Np}$  from the ceramic waste form at the end of the same time period is only about 75% of the corresponding release of  $^{129}\text{I}$  because the half-life for  $^{237}\text{Np}$  is nearly a factor of ten shorter than the half-life for  $^{129}\text{I}$ .

## V. CONCLUSIONS

The normalized cumulative release curves show that with the current model, the normalized release from the Yucca Mountain engineered barrier system for the metal waste form can be expected to be much lower than that from DOE spent nuclear fuel at all times and close to or lower than that for DHLW glass for the first 30,000 years after emplacement in the repository. With the current model, the normalized release of  $^{129}\text{I}$  from the ceramic waste form is lower than that from DHLW glass at all times and compares favorably with cladded commercial spent nuclear fuel for much of the first 20,000 years. The initial inventory of the ceramic waste form does not include  $^{99}\text{Tc}$ . The comparison for  $^{237}\text{Np}$  shows that the ceramic

waste form also has a lower normalized release than DHLW at all times after about the first 20,000 years. Prior to 20,000 years, solubility considerations in combination with a small  $^{237}\text{Np}$  inventory in the ANL ceramic waste form packages cause the DHLW glass to have a lower normalized cumulative release. The lower cracking factor is responsible for the good performance of the ceramic waste form. This factor is not well known for either the ceramic or the defense high-level waste forms. As noted earlier, the cracking factor for the ceramic waste form is based on measurement, but the measurement was for a smaller sized HIP can than is planned for the repository. In spite of uncertainties in the cracking factor and in other aspects of the ceramic and metal waste form degradation modeling, the normalized cumulative release curves show that the retention of radionuclides by the ANL waste forms will be similar to and in some cases better than the retention anticipated for the more robust waste forms currently planned for disposal in the repository.

The waste-form degradation models used for the analyses described are preliminary and are based on a limited amount of experimental data. More data have become available since these analyses were performed and updated models are being developed. The calculations described here will be repeated as appropriate when the updated models become available.

## ACKNOWLEDGMENTS

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