RADIONUCLIDE RELEASE RATES AT THE RECEPTOR LOCATION FOR THE PROPOSED HIGH-LEVEL RADIOACTIVE WASTE DISPOSAL SITE AT YUCCA MOUNTAIN

S. MOHANTY*, R.W. RICE**
***Southwest Research Institute, CNWRA, 6220 Culebra Rd, San Antonio, TX 78228, smohanty@swri.edu**
****Consultant, El Paso, TX, 79922, rwrice@aol.com**

ABSTRACT

The Nuclear Regulatory Commission (NRC) Total-system Performance Assessment (TPA) code is a tool to independently evaluate the long-term performance of the proposed Yucca Mountain (YM) high-level radioactive waste (HLW) repository by modeling processes such as the dissolution of the spent nuclear fuel (SNF) and the subsequent transport through unsaturated and saturated highly heterogeneous fractured porous media to a hypothetical receptor location. The release rates of radionuclides at the receptor location corresponding to some of the SNF dissolution models show a sinusoidal trend with an overall decrease in the rate with time; for other models the sinusoidal behavior is nonexistent. This study attributes these trends to mechanisms related to radionuclide-specific sorption properties in the saturated zone alluvium and the spatial discretization of the repository into subareas. Other mechanisms which might affect the release rates, such as solubility limits, radioactive decay and ingrowth, inventory depletion, and transport properties of the unsaturated zone (UZ), do not significantly contribute to these trends.

INTRODUCTION

The NRC has the responsibility to review the license application for the HLW repository site at YM. In support of its regulatory review activities, the NRC staff has focused on detailed technical evaluation [1,2] to understand and quantify the isolation characteristics and capabilities of the proposed YM repository system. To support these technical assessments, the NRC and the Center for Nuclear Waste Regulatory Analyses (CNWRA) recently developed Version 3 of the TPA code [3].

YM is located in a semi-arid environment on the Nevada Test Site in southern Nevada, and rises several hundred meters above the surrounding land. The current design for the YM repository is to dispose of SNF in waste packages (WPs) emplaced in drifts approximately 300 m below the top of YM and 300 m above the water table. Following emplacement in the repository, the WPs will eventually fail, and the infiltrating water will contact the SNF in the WP and cause SNF dissolution. After the SNF dissolves into the contacting water, the flowing water will transport radionuclides out of the engineered barrier system (EBS), which is comprised of the WP, concrete invert that supports the WP, and possibly shields and backfill, through the hydrologically UZ and saturated zone (SZ) to a hypothetical receptor group located 20 km from the repository footprint.

Because of the high level of uncertainty in projecting the probable evolution of the repository, alternative conceptual models are used to evaluate the performance of the YM repository. To represent uncertainty in predicting the SNF dissolution rate, which is one of several key factors in assessing repository performance, four alternative conceptual models have

been proposed [3] to characterize the nature of the time evolution of releases from the EBS. While the EBS releases show an expected decreasing trend for each of the SNF dissolution rate models, a very distinctive, yet peculiar sinusoidal behavior is observed in the calculational results of radionuclide release rates at the receptor location. To investigate this behavior, this paper first describes the general trends in the receptor location release rates and then hypothesizes several potential mechanisms. These mechanisms include: (i) solubility limits of the radionuclides, (ii) decay and ingrowth of the radionuclide inventory, (iii) inventory depletion of the SNF in the WP, (iv) transport properties of the UZ and SZ, and (v) spatial discretization of the repository. The paper guides the reader through contributions from these mechanisms to the trends exhibited in the receptor location release rates.

MODEL DESCRIPTION

To analyze the behavior of the receptor location release rates, the repository is discretized into seven subareas representing different regions in the repository that are comprised of different thicknesses of hydrostratigraphic units, chemical sorption (K_d) and hydrologic properties, and matrix/fracture flow interactions. For each subarea, the transport of radionuclides through the UZ and SZ is modeled with a vertical column, representing the UZ, connected to a horizontal streamtube, representing the SZ. Simulations are conducted to 100,000 yr.

For the release and transport calculations, eleven radionuclides in the SNF are modeled: Am-241, C-14, Cl-36, Cm-245, I-129, Np-237, Pu-239, Se-79, Tc-99, Th-230, and U-234. These radionuclides were identified in a screening process as having the potential to significantly contribute to dose at the receptor location [4]. Two decay chains are also modeled: Cm-245–>Am-241–>Np-237 and U-234–>Th-230. The EBS release rates of these radionuclides provide a source term or time-dependent boundary condition for the UZ and SZ transport model. Using this source term, the release rates are computed at the outlet of the UZ and SZ flow path for each subarea. The outlet of the SZ flow paths corresponds to the location of the pumping well used by the receptor group.

The four SNF dissolution models used to compute the source term for UZ and SZ transport represent alternative conceptualizations of SNF release rates from the WP and are expected to bound the conceptual uncertainties associated with conditions that may exist in and around the WP and SNF subsequent to WP failure [3]. Model 1 estimates the SNF dissolution rate in solutions containing carbonate anions. The SNF dissolution rate in the presence of Ca and Si ions, which are found in the YM groundwater, is calculated in Model 2. The user has the option of specifying a SNF dissolution rate in Model 3, such as from a natural analog model. The formation of secondary minerals is considered in Model 4. Mathematical formulations of the SNF dissolution models and the associated assumptions are given elsewhere [4] and are not the focus of this paper. Rather, the modifications to the trends in the release rates at the EBS due to the transport of radionuclides through the UZ and SZ is the primary focus.

RESULTS AND DISCUSSION

The TPA simulations are conducted by exercising the four SNF dissolution rate models first to determine the release rates from the EBS. The release rate then is used as a boundary condition for transport through the UZ and SZ. The discussion describes detailed analyses of the results which provide an explanation for the release rate trends based on general factors, such as radionuclide and subarea effects, and the proposed mechanisms identified previously.

The release rates for I-129, Tc-99, Se-79, and Cl-36, which represent four of the eleven radionuclides evaluated in TPA simulations, are provided in this section. These four radionuclides are analyzed in detail because they have the largest contribution to dose [4].

The total release rates for these four radionuclides out of the EBS and at the receptor location calculated with the four SNF dissolution rate models are provided in figures 1(a) and 1(b), respectively. Figure 1(a) shows two peaks which correspond to two distinct WP failure times, and the height of each peak is proportional to the number of failed WPs. The peak release rate, which occurs before 5,000 yr, is attributable to initially failed WPs (i.e., defective or damaged prior to or during emplacement), while the releases at about 17,000 yr are from WPs failed by corrosion.

For discussion purposes, the four SNF dissolution rate models can be classified into two categories, Group1 and Group 2, where Group 1 models show undulated release rates with an overall decreasing trend in rate with time, and Group 2 models show rather smooth release rate characteristics. As shown in figure 1(a), the EBS release rates for the Group 1 models, Models 1 and 4 that exhibit fast and slow release rates, feature an increase to a peak value and thereafter decrease several orders of magnitude. The EBS release rates for the Group 2 models, Models 2 and 3 that have an intermediate release rate, reach a maximum value that remains relatively constant with time. The rapid decrease in release rates for Group 1 models is attributable to source depletion for Model 1 and to instantaneous releases from the gap inventory (i.e., portion of the radionuclide inventory assumed to be held loosely on the grain boundaries, cladding/fuel gap, and cladding) for Model 4, whereas the Group 2 models maintain an intermediate release rate which is not large enough to be dominated by source depletion or small enough to be dominated by instantaneous releases from the gap inventory.

To investigate the trends in the Group1 and Group 2 models, particularly the undulations and decrease in the overall trend in the release rates after 50,000 yr for the Group 1 models, receptor location release rates computed using Models 1 and 2 are provided in figure 2 for each radionuclide. Only the results from Models 1 and 2 are presented because the trends in their release rates are representative of the Group 1 and Group 2 models during the 100,000-yr simulation period. The total release in figure 2(a) for Model 1 and in figure 2(b) for Model 2 corresponds to the release rates provided in figure 1(b). Results in figure 2 show a rapid decrease in release rates for individual radionuclides for the Group 1 models compared to the Group 2 models. This rapid decrease is responsible for the decrease in the release rates shown in figure 1(b) after 50,000 yr. Additionally, the results in figure 2 illustrate that different radionuclides contribute to the undulations in the total release. The effect is more evident in figure 2(a), which shows that at early times the total release is attributable to Cl-36 and I-129, and at later times to Tc-99 and Se-79. However, although figure 2 shows the effect of different radionuclides on the release rate, these figures do not indicate the reasons for undulations in the release rate for the same radionuclides, such as Tc-99.

To further explore the reasons for the undulations in figure 2, Tc-99 release rates at the receptor location are presented in figure 3 for each subarea. In this figure, the Tc-99 release rates are presented, because the trends in the Tc-99 release rates are representative of the trends in the I-129, Se-79, and Cl-36 release rates. The total release of Tc-99 in figure 3(a) for Model 1 and in figure 3(b) for Model 2 correspond to the Tc-99 release rates in figures 2(a) and 2(b), respectively. From figure 3, it is noticeable that the undulations in the Tc-99 release rate is associated with contributions from different subareas.

Thus, the results in figure 2 and 3 identify radionuclide and subarea contributions to the undulations in the release rates and the causes for the rapid decrease observed in figure 1(b) after

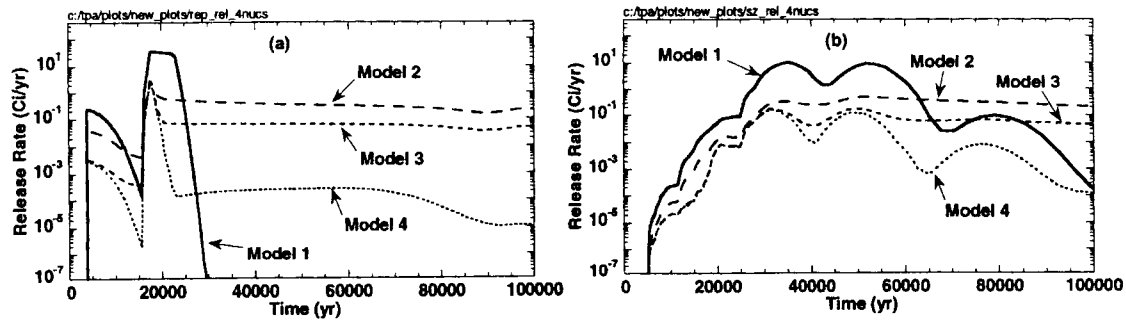

Figure 1. Total release rates for Tc-99, I-129, Se-79, and Cl-36 (a) out of the EBS and (b) at the receptor location for the four SNF dissolution rate models

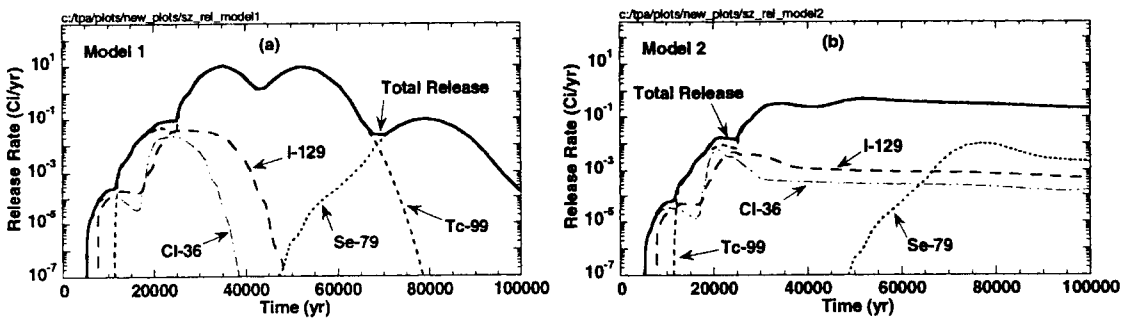

Figure 2. Release rates for Tc-99, I-129, Se-79, and Cl-36 at the receptor location using (a) Group 1 (Model 1) and (b) Group 2 (Model 2) SNF dissolution rate models

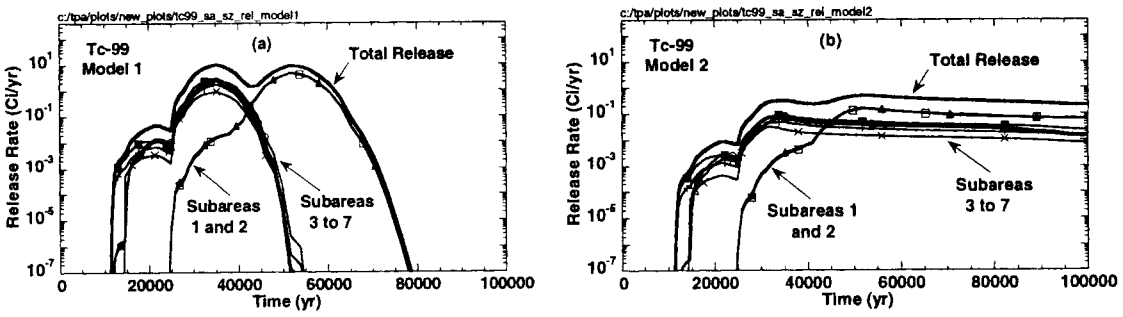

Figure 3. Release rates for Tc-99 from each subarea at the receptor location using (a) Group 1 (Model 1) and (b) Group 2 (Model 2) SNF dissolution rate models

50,000 yr. The mechanisms proposed previously in this paper are associated with these trends. The following sections discuss the impacts of these proposed mechanisms on the receptor location release rates.

Solubility Limits

The solubility of a radionuclide can affect the magnitude and time of the peak EBS release rate, which may change the release rate at the receptor location. Releases that are not solubility limited are expected to exhibit a more rapid increase in the release rate compared to releases that are solubility limited which show a more gradual increase in the release rate. However, the radionuclides I-129, Tc-99, Se-79, and Cl-36 are highly soluble [3] and the only significant difference in the EBS release rates, which were analyzed but not included in this paper, is the magnitude of the releases for the Group 1 and Group 2 models. This difference is caused by the initial inventory of SNF in the WP. Consequently, solubilities do not cause the difference between the release rates for the Group 1 and Group 2 models.

Radioactive Decay and Ingrowth

The release rate of a radionuclide may be affected by radioactive decay and ingrowth. The radionuclides I-129, Tc-99, Se-79, and Cl-36 are not modeled in the TPA code with chain decay. Because the half-lives of these radionuclides, 1.57×10^7, 2.13×10^5, 6.5×10^4, 3.01×10^5 yr, are the same order of magnitude as the 100,000-yr simulation time, half-lives do not significantly decrease the inventory available for release, and thus the EBS release rates, by orders of magnitude as shown in the Group 1 model release rates in figure 1(b). Consequently, radioactive decay and ingrowth do not cause the differences observed in the Group 1 and Group 2 models in figure 1(b).

Inventory Depletion

A depletion of inventory may cause a significant change in the release rate with time because the amount of a radionuclide released from the SNF is related to the amount available for release. Except for Model 1 which quickly releases the WP inventory, every SNF dissolution model shows a relatively constant release rate with time after the gap inventory is released. Thus, inventory depletion is not a primary cause of the behavior of the Group 1 models in figure 1(b).

Transport Properties of the Unsaturated and Saturated Zones

The transport of radionuclides to the receptor location may be delayed by sorption and desorption in the UZ and SZ. However, in a separate analysis of results not presented in this paper, the UZ release rates are not significantly different from the EBS release rates. Thus, the analysis presented in this section will focus on transport in the SZ.

Figure 2 displays radionuclide-specific differences in the receptor location release rates. For the total release, Cl-36 has the largest contribution prior to about 8,000 yr. From about 8,000 to 12,000 yr, both Cl-36 and I-129 contribute the most to total release. Between about 12,000 and 68,000 yr, Tc-99 dominates total release and, thereafter, Se-79 is the major contributor to total release. The differences in the arrival time of the peak release for these radionuclides are caused by retardation in the SZ alluvium. Radionuclides with the smallest retardation factors arrive

earlier at the receptor location, while later arrival times correspond to radionuclides with larger retardation factors. The SZ alluvium retardation factors are 1.0, 2.0, 5.5, and 22.4 for Cl-36, I-129, Tc-99, and Se-79, respectively [3]. Thus, for the Group 1 models, the sinusoidal behavior observed in figure 1(b) can be attributed to contributions from these various radionuclides. Sorption in the SZ alluvium does not cause a similar trend for the Group 2 models because the EBS release rates, as shown in figure 1(a), remain relatively constant.

Spatial Discretization

The EBS release rates are computed for seven subareas in the TPA code and each subarea has a unique UZ and SZ hydrostratigraphy that could affect radionuclide transport. The UZ is previously identified as having no effect on the receptor location release rates. Therefore, only the effects of spatial discretization in the SZ will be discussed.

The Tc-99 release rates discussed earlier and provided in figure 3 for the Group 1 and Group 2 models show the same behavior. The releases from subarea 1 and 2 are delayed by about 15,000 yr when compared to the release rates from subareas 3 through 7. This delay causes two peak release rates at about 30,000 and 50,000 yr in figure 1(b). These peaks are attributable to the differences in the length of the SZ flow path and the groundwater travel time. For subareas 1 and 2, the SZ flow path length is approximately 4,000 to 7,000 m greater than the length of the SZ flow path for subareas 3 through 7 [3]. The differences in the flow path length produce an SZ groundwater travel time of about 6,000 yr for subareas 1 and 2, compared to approximately 2,800 yr for subareas 3 through 7. Consequently, spatial discretization of the repository into subareas is also responsible for the sinusoidal behavior of the Group 1 models.

SUMMARY AND CONCLUSIONS

Two categories of SNF dissolution rate models were identified in this paper: Group 1 models which exhibit release rates dominated by source depletion and instantaneous releases from the gap inventory, and Group 2 models which show release rates that are not dominated by these effects and have rather smooth release rate characteristics. The release rates at the receptor location computed using the Group 1 models show a sinusoidal nature and a relatively rapid decrease after 50,000 yr while, for the Group 2 models, the sinusoidal behavior vanishes. This paper proposes that these trends in the release rate may be caused by the effects of solubility limits, inventory depletion, radioactive decay and ingrowth, UZ and SZ transport properties, and repository spatial discretization. After analyzing the EBS and receptor location release rates, an explanation for the trends in the receptor location release rates was identified.

The sinusoidal trend in the receptor location release rates calculated using Group 1 models may be attributed to differences in (1) SZ alluvium sorption for each radionuclide, and (2) spatial dependencies related to the length of the SZ flow path for each subarea. Furthermore, the decrease in the receptor location release rates for the Group 1 models after 50,000 yr is caused by the rapid decrease in the EBS release rate. The Group 2 models do not exhibit these trends, because the receptor location release rates are relatively constant with time.

Thus, the SNF dissolution rate model utilized in a TPA code simulation can strongly affect the trend in the release rates at the receptor location. The behavior of the receptor location release rates is important, because these release rates are used to compute dose to the receptor, which is the performance measure for the HLW repository at YM.

ACKNOWLEDGMENTS

The paper was prepared to document work performed by the Center for Nuclear Waste Regulatory Analyses (CNWRA) for the Nuclear Regulatory Commission (NRC) under contract No. NRC–02–97–009. The activity reported here was performed on behalf of the Office of Nuclear Material Safety and Safeguards. The paper is an independent product of the CNWRA and does not necessarily reflect the views or regulatory position of the NRC.

REFERENCES

1. R.B. Codell, N. Eisenberg, D. Fehringer, W. Ford, T. Margulies, T. McCartin, J. Park, and J. Randall, *Initial Demonstration of the NRC's Capability to Conduct a Performance Assessment for a High-Level Waste Repository*, NUREG–1327, Nuclear Regulatory Commission, Washington, DC, 1992.

2. R.G. Wescott, M.P. Lee, N.A. Eisenberg, T.J. McCartin, and R.G. Baca, eds., *NRC Iterative Performance Assessment Phase 2*, NUREG–1464, Nuclear Regulatory Commission, Washington, DC, 1995.

3. S. Mohanty and T.J. McCartin, *Total-system Performance Assessment (TPA) Version 3.2 Code: Module Description and User's Guide*, Center for Nuclear Waste Regulatory Analyses, San Antonio, TX, 1998.

4. S. Mohanty, R. Codell, R.W. Rice, J. Weldy, Y. Lu, R.M. Byrne, T.J. McCartin, M.S. Jarzemba, and G.W. Wittmeyer, *System-level Repository Sensitivity Analysis Using TPA Version 3.2 Code*, Center for Nuclear Waste Regulatory Analyses, San Antonio, TX, 1999.