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COUPLED PROCESS MODELING AND WASTE PACKAGE PERFORMANCE

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The interaction of borosilicate waste glasses with water has been studied extensively and reasonably good models are available that describe the reaction kinetics and solution chemical effects. Unfortunately, these models have not been utilized in performance assessment analyses, except in estimating radionuclide solubilities at the waste form surface. A geochemical model has been incorporated in the AREST code to examine the coupled processes of glass dissolution and transport within the engineered barrier system. Our calculations show that the typical assumptions used in performance assessment analyses, such as fixed solubilities or constant reaction rate at the waste form surface, do not always give conservative or realistic predictions of radionuclide release. Varying the transport properties of the waste package materials is shown to give counterintuitive effects on the release rates of some radionuclides. The use of noncoupled performance assessment models could lead a repository designer to an erroneous conclusion regarding the relative benefit of one waste package design or host rock setting over another.

Coupled Process Modeling and Waste Package Performance

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INTRODUCTION

Current performance assessment models for the engineered-barrier system utilize a fixed solubility limit or constant dissolution rate of the waste form as conservative boundary conditions for near-field transport calculations. The transport equations can then be solved either analytically[1] or numerically.[2] With these models, changing the transport properties of the system, such as the diffusion coefficient, tortuosity, or porosity, affects radionuclide release rates in a straightforward manner consistent with the structure of the governing transport equations and has no affect on the assumed solubilities or dissolution rate of the waste form. With a glass waste form, special problems arise in applying these models because the glass is a metastable solid that cannot be in equilibrium with an aqueous solution. Hence the use of equilibrium thermodynamics to calculate a fixed solubility for use in performance assessment studies is inherently problematic. In this paper, we present results from a geochemical model, that has been implemented in the Analytical Repository Source-Term (AREST) code[3,4], that more appropriately describes the coupling between glass/water chemical reactions and transport across the engineered-barrier system.

MODEL DESCRIPTION

The AREST code is intended to provide a quantitative probabilistic assessment of the performance of the individual barriers of the overall engineered barrier system (EBS). AREST contains three major components: 1) waste package containment (WPC), 2) waste package release (WPR) , and 3) engineered system release (ESR).

The WPC models the effective life of a waste container using a statistical distribution of failures. After containment failure, the waste package release model describes the radionuclide release from the waste form and the migration outward through the waste package. AREST contains several analytical models for estimating release from the waste package. There also exists a numerical release model, in AREST, which estimates the diffusive migration through the backfill and diffusion/advection into the host rock. AREST estimates WPC and WPR for each simulated waste package. Individual release estimates are then combined by the engineered system release model to produce estimates of total release.

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A geochemically-based model for glass dissolution[5,6] has been incorporated into AREST. This model considers the heterogeneous dissolution of a glass in an open system that can also include other EBS materials. A mass balance is developed to calculate the time rate of change in the extent of the reaction progress variable for irreversible waste form dissolution.[7] Mineral affinities and elemental concentrations as a function of reaction progress, pH, and Eh are calculated with the help of the EQ3/6 code before running AREST.

RESULTS AND CONCLUSIONS

Figure 1 shows the results of a release rate prediction for a hypothetical waste package surrounded by a 1-m thick clay backfill assuming a constant solubility for both Am and Pu at the waste form surface. The release rates for these two actinides drop by over two orders of magnitude by decreasing the diffusion coefficient in the clay by a factor of 10. This behavior is consistent with expectations from simple mass transfer theory[1] where chemical changes in the waste package environment are ignored.

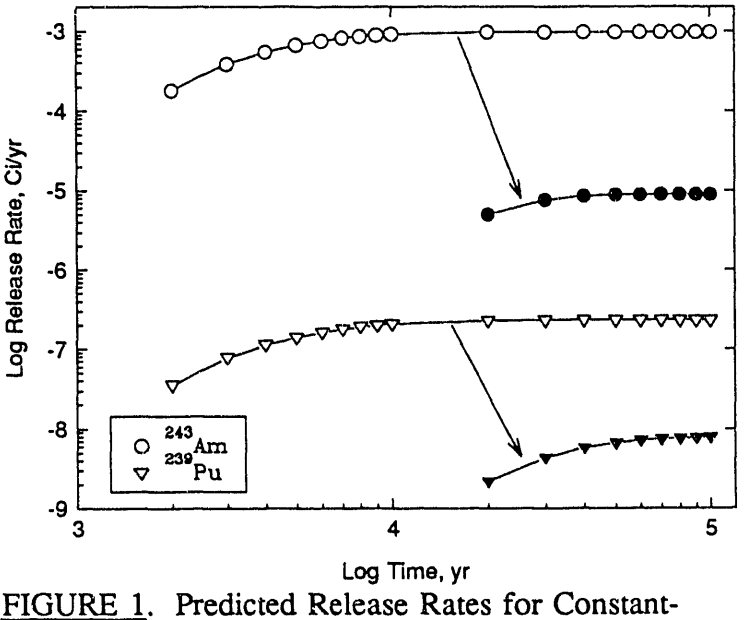

FIGURE 1. Predicted Release Rates for Constant-Solubility Release Model. Open symbols: $D=10^{-5}$ cm^2/s; Solid symbols: $D=10^{-6}$ cm^2/s.

Implementation of a coupled model in the AREST code that accounts for the chemical effects of glass waste form dissolution significantly affects the release rate predictions for this waste package design, as illustrated in Figure 2. The interaction of the glass waste form with the groundwater increases both the pH and Eh. For Am, the increasing pH increases the solubility of $AmOHCO_3(s)$, although the change is small (less than a factor of 2). Consequently, ^{243}Am release rates decrease with a decrease in diffusion coefficient from 10^{-5} to 10^{-6} cm^2/s as was found for the constant solubility model. However, for Pu, the Eh changes result in several orders of magnitude increase in the solubility of $PuO_2(s)$.[b] As a result, the predicted release rate for ^{239}Pu increases with the decrease in assumed diffusion coefficient. These results have important implications for repository designers using simple models for waste performance assessment. As illustrated in Figure 1, the noncoupled models can lead to an inaccurate conclusion regarding the performance benefit of one waste package

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[b]PuO_2 was the stable solid phase predicted by the EQ3/6 simulations.

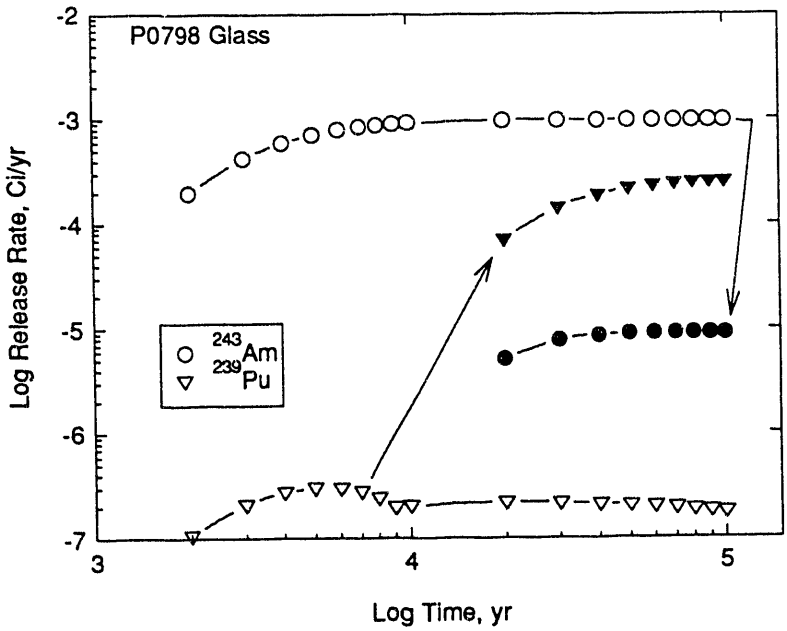

FIGURE 2. Predicted Release Rates Using Coupled Release Model. Open symbols: $D=10^{-5}$ cm^2/s; Solid symbols: $D=10^{-6}$ cm^2/s.

design or host rock setting over another. The typical analysis for release from the engineered barrier system using a constant concentration boundary condition at the waste form surface does not necessarily produce realistic or conservative results.

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