CONF-960911--3

# SENSITIVITY OF THE ENGINEERED BARRIER SYSTEM (EBS) RELEASE RATE TO ALTERNATIVE CONCEPTUAL MODELS OF ADVECTIVE RELEASE FROM WASTE PACKAGES UNDER DRIPPING FRACTURES

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## ABSTRACT

Simulations were conducted to analyze the sensitivity of the engineered barrier system (EBS) release rate to alternative conceptual models of the advective release from waste packages under dripping fractures. The first conceptual model assumed that dripping water directly contacts the waste form inside the "failed" waste package, and radionuclides are released from the EBS by advection. The second conceptual model assumed that dripping water is diverted around the "failed" waste package (because of the presence of corrosion products plugging the perforations) and dripping water is prevented from directly contacting the waste form. In the second model, radionuclides were assumed to transport through the perforations by diffusion, and, once outside the waste package, to be released from the EBS by advection. The second model was to incorporate more realism into the EBS release calculations.

For the case with the second EBS release model, most radionuclides had significantly lower peak EBS release rates (from at least one to several orders of magnitude) than with the first EBS release model. The impacts of the alternative EBS release models were greater for the radionuclides with a low solubility (or solubility-limited radionuclides) than for the radionuclides with a high solubility (or waste form dissolution-limited radionuclides). The analyses indicated that the EBS release model representating advection through a "failed" waste package (the first EBS release model) may be too conservative in predicting the EBS performance. One major implication from this sensitivity study was that a "failed" waste package container with multiple perforations may still be able to perform effectively as an important barrier to radionuclide release.

## INTRODUCTION

A potential repository at Yucca Mountain, Nevada, USA, is being studied for the disposal of the nation's high-level nuclear waste including spent nuclear fuel (SF) and vitrified defense high-level waste (DHLW). If found suitable, the repository will be constructed in an unsaturated geologic formation below the mountain. In the current design concept, a robust engineered barrier system (EBS) is employed. The primary component of the EBS is a multi-barrier waste package container, which will be designed to meet the regulatory requirements (currently, 10 CFR 60) for substantially complete containment of the nuclear waste and





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controlled release upon failure of the waste container.

In the potential repository, the decay heat from the wastes (mostly from spent nuclear fuel) is expected to drive off moisture from the vicinity of the waste packages. The combined effects of this decay heat and the unsaturated condition would be likely to create dry and hot near-field conditions for an extended period of time and provide a prolonged waste package service life. During the waste containment period, some waste packages may be exposed to dripping fractures with local and/or sporadic groundwater dripping. Besides enhancing the waste package container corrosion, the dripping fractures may contribute to release of radionuclide by advection from the failed waste packages, hence considerably increasing the release rates over diffusion-only release. This paper discusses the study results on the sensitivity of the EBS release rates of radionuclides to two alternative EBS advective release conceptual models, which were studied as part of the recent iteration of the total system performance assessment (TSPA-1995) for the potential repository [1].

## DESCRIPTION OF ALTERNATIVE EBS ADVECTIVE RELEASE MODELS

The following two EBS advective transport conceptual models were developed and incorporated to investigate their effects on the waste package and EBS release rates. Brief discussions of each conceptual model are given below.

### EBS Advective Release Conceptual Model I (Drip-On-Waste-Form Model)

In this model, once a waste package fails by pitting corrosion (i.e., having at least one pit penetration through the waste package container), the entire waste form was assumed to be exposed to the (dripping) advective flux. The flux was assumed to directly contact the waste form, and the mobilized radionuclides were released by advection. A schematic of this conceptual model is shown in Figure 1. The advective release rate is calculated using the following conventional advective release rate equation

$$\dot{M}_{EBS}^{ad} = Q_{drip}\, C_0 \qquad (1)$$

where $\dot{M}_{EBS}^{ad}$ is the advective mass transfer rate from the EBS (moles/yr), $Q_{drip}$ is the volumetric dripping flow rate on the waste package ($m^3$/yr), and $C_0$ is the radionuclide concentration at the waste form surface (moles/$m^3$). The diffusive release from the "failed" waste package under dripping fractures was also included in this model [2]. The total release was then the sum of the advective and diffusive releases, and the advective release component was always dominant. For "failed" waste packages that were not subjected to dripping water, radionuclides were assumed to be released by diffusion only, and the diffusion coefficient was determined as a function of the volumetric water content of the crushed tuff gravel invert underneath the waste package [2].

### EBS Advective Release Conceptual Model II (Drip-On-Waste-Package Model)

EBS Release Model I (discussed above) is conservative in that it does not account for any potential performance credit for the "partially" failed (or perforated) waste container as a barrier to radionuclide release. However, it is expected that the perforations in the failed

waste containers are filled with fine, gel-like porous corrosion products which may keep dripping groundwater from flowing through the waste package and thus from directly contacting the waste form. Therefore, in this conceptual model (drip-on-waste-package model), dripping groundwater was assumed to be diverted around, not through, the waste package container. Radionuclides were assumed to be transported through the perforations (filled with the corrosion-products) by diffusion to the outside of the waste container, and, once outside, these radionuclides were released from the EBS by advection through the underlying invert and to the edge of the EBS. Figure 2 shows a schematic of this second advective release conceptual model (i.e., drip-on-waste-package model). If steady-state diffusion through the perforations in the waste package container is assumed, then the EBS advective release rate from a waste package container with $N$ uniformly distributed perforations can be expressed as follows

$$\dot{M}_{EBS}^{ad} = \frac{Q_{drip} C_0}{\frac{1}{N\pi r^2}\left[\frac{\pi r}{4}\left(\frac{1}{D_0\varepsilon_0}+\frac{1}{D_2\varepsilon_2}\right)+\frac{l}{D_1\varepsilon_1}\right]Q_{drip}+1} \qquad (2)$$

where $r$ and $l$ are the radius and length, respectively, of the cylindrical perforation. $\varepsilon_i$ and $D_i$ are the porosity and diffusion coefficient, respectively, in the region $i$ ($i$ = 0, 1, and 2): Region 0 represents the inner surface of the waste container, Region 1 is the perforation filled with porous corrosion products, and Region 2 represents the outer surface of the waste container. The porosity just inside and outside the waste container ($\varepsilon_0$ and $\varepsilon_2$) was assumed to be 1.0, and $D_0$ and $D_2$ were assumed to be $10^{-5}$ cm$^2$/sec. The porosity of the corrosion products ($\varepsilon_1$) was assumed 0.4, and the diffusion coefficient in the perforation ($D_1$) was determined as a function of the volumetric water content of the corrosion products. Details for the derivation of Equation (2) are given by Lee, et al [2,3]. As can be seen in Equation (2), important parameters in this model that control the advective release rate are the volumetric dripping flow rate ($Q_{drip}$), the number of perforations ($N$), radius of the perforation ($r$), and thickness of the container wall ($l$). This model incorporates more realism into the EBS release calculation because it assumes the advective release rate from a waste container depends on the number of pit penetrations at a given time. As in the first EBS release conceptual model, radionuclides were released by diffusion only from "failed" waste packages that were not subjected to dripping fractures [2].

## ABSTRACTIONS OF PROCESS-LEVEL MODEL RESULTS

Repository Integration Program (RIP) [4], a total system performance assessment (TSPA) model, was used to simulate the EBS performance. This TSPA model does not include detailed process-level models, rather it requires abstractions from the process-level models to capture important process behaviors in a simple functional form, a probabilistic manner, or a combination of both. The following are brief descriptions of major abstractions that were implemented into RIP for the EBS performance analyses. Details are found in the recent TSPA-1995 report [1].

(1) The near-field conditions (i.e., temperature, relative humidity and liquid saturation) as a function of time were simulated with a drift-scale thermal-hydrologic model [5], and

their results were abstracted.

(2) The failure and (upon failure) degradation histories of waste packages as a function of time (subjected to the near-field conditions) were developed using a stochastic waste package degradation model which was developed as part of TSPA-1995 [6]. The results were abstracted in such a way that the waste packages were grouped into 6 groups by their failure time with an equal number of waste packages in each group. The same grouping procedure was used for each waste form type (giving a total of 12 groups). For each group, an average failure time and a representative pitting degradation history as a function of time were developed, and all the waste packages in each group were assumed to have the same (average) failure time and the same (representative) degradation history. This abstraction provides the number of pit penetrations ($N$ in equation (2)) in the waste packages as a function time.

(3) The drift-scale hydrology profiles in terms of matrix flow and fracture flow were abstracted in such a way that the number of waste packages under dripping fractures were estimated as a function of the spatial distribution of the repository infiltration flux and the saturated matrix hydraulic conductivity of the near-field rock surrounding the repository drifts. In the abstraction, the repository infiltration rate was sampled from two distributions: 1) low infiltration rate with 0.01-0.05 mm/yr (uniformly distributed); and 2) high infiltration rate with 0.5-2.0 mm/yr (uniformly distributed). This abstraction provides $Q_{drip}$ in equations (1) and (2).

(4) Representative compositions and surface areas of the waste forms (spent nuclear fuel and defense high-level glass) were developed.

(5) Dissolution rates of the waste forms were abstracted as a function of temperature, pH, and total carbonate concentration (the last for spent fuel only).

(6) Mathematical models for the diffusive and advective EBS release rates from a waste package container with multiple perforations were developed and implemented [2,3].

## MAJOR ASSUMPTIONS AND KEY PARAMETER VALUES

Discussed in this section are: 1) the major assumptions incorporated into the RIP simulations for the waste package and EBS subsystem performance analyses, and 2) the key parameter values used in the simulations.

(1) The effective catchment area for a waste package in an emplacement drift was assumed to be four times the length times the diameter of the waste package (i.e., four times the waste-package maximum cross-sectional area perpendicular to flow). The use of the effective catchment area was to incorporate the uncertainty concerning how fracture flow will be distributed once it intersects the drift. It was assumed that the advective flux entering into the effective catchment area is "focused" onto the waste package.

(2) The waste package containers for both spent fuel and defense high-level glass were assumed to have the same two-layer container design with a 10-cm thick carbon steel outer layer and a 2-cm thick Alloy 825 inner layer. Although the carbon steel outer layer would be thinned due to wastage from general corrosion, the waste container was assumed to have a constant wall thickness of 12 cm ($l$ in equation (2)) for the entire simulation period.

(3) When the waste package container had at least one pit penetration (or waste package

"failure") and the surface temperature was below 100 °C, the entire waste form surface was assumed to be covered with a "thin" water film (with a uniform thickness of 1.0 mm). Alteration/dissolution of the waste form was assumed to be initiated immediately after the first pit penetration. The water film thickness was used in the calculation of the radionuclide concentration at the waste form surface.

(4) The releases of $^{14}C$, $^{36}Cl$, and $^{129}I$ from the EBS were assumed to be gaseous. The gaseous elements were assumed to escape unimpeded from the EBS, and then dissolve into the aqueous phase and transport to the geosphere. However, there is some uncertainty regarding the dominant release behavior of $^{129}I$ from the waste package and EBS (i.e., gaseous and aqueous). A potential loss of iodine as $I_2$ gas from an aqueous solution was indicated in a recent study [7].

(5) If a "failed" waste package was under dripping fractures, radionuclides were assumed to be released from the EBS both by advection and diffusion. In this case the diffusion coefficient in the perforation ($D_1$) in equation (2) was set to $10^{-7}$ $cm^2/sec$ [8]. In the absence of dripping water, a diffusive release was assumed, and the diffusion coefficient was calculated using an empirical functional form as a function of the volumetric water content [1,2].

Waste package parameters used in the RIP simulations are given in Table 1. Radionuclides with gap and grain-boundary inventories were $^{14}C$, $^{135}Cs$, $^{129}I$, $^{79}Se$ and $^{99}Tc$, and their inventories were assumed to be 2% of their individual total inventories, except for $^{14}C$ whose inventory was assumed to have a range uniformly distributed between 1.25 and 5.75%. The surface area assumed for spent fuel was 3.815 $cm^2$/(cm matrix), which was converted to $5.57 \times 10^{-5}$ $m^2/g$ using the spent fuel pellet diameter of 0.93 cm and the density of 10.08 $g/cm^3$ [9]. The surface area for defense high-level waste glass was assumed to be in a range uniformly distributed between 50 and 150 $m^2$ per pour canister (each waste package contains four pour canisters). Detailed discussion of the key parameters is given in the recent TSPA-1995 report [1].

## RESULTS AND DISCUSSION

The RIP simulations for the analysis of the sensitivity of the EBS release rates to the alternative advective release models (EBS Release Models I and II) were conducted for the case with 83 metric tons of uranium (MTU)/acre thermal loading, no backfill and high infiltration. The abstractions for waste package degradation implemented in the simulations were for the case in which corrosion of the outer barrier (carbon steel) is initiated when temperature is below 100°C and relative humidity is above some threshold between 65 and 75%. The simulations were performed for 10,000 years using the expected values of various stochastic process parameters (i.e., waste form dissolution rate, radionuclide solubility, infiltration rate, etc.).

The EBS release rates of the gaseous release radionuclides ($^{14}C$, $^{36}Cl$ and $^{129}I$) were not affected by the alternative EBS release models, thus they are not included in this paper. As expected, the analyses showed that the conservative EBS release model (EBS Release Model I or Drip-on-Waste-Form Model) yielded higher peak EBS release rates for all the radionuclides (including those not shown in this paper) than EBS Release Model II. Figures 3 and 4 show the results for $^{99}Tc$ and $^{135}Cs$, respectively, which have a substantial gap and

grain-boundary inventory (2% for each), are soluble in water (a median solubility of $10^{-3}$ M for $^{99}$Tc and $2.9 \times 10^{-3}$ M for $^{135}$Cs) and mobile, and have a long half-life ($2.12 \times 10^5$ years for $^{99}$Tc and $3 \times 10^6$ years for $^{135}$Cs). Each of the five "peaks" shown with the EBS Release Model I in the figures corresponds to the failures of the waste packages in the first five waste package groups and are "artifacts" of the waste package abstraction process (i.e., all the waste packages in each group fail at the same "average" failure time for that group). In general, the shape of the EBS release rate curves with the EBS Release Model II (Drip-on-Waste-Package Model) mimics the waste package pitting degradation histories [1,2,6].

As shown in Figure 3, the peak EBS release rate for $^{99}$Tc with EBS Release Model II (about 12 Ci/yr at 10,000 years) is about an order of magnitude lower than the peak release rate with EBS Release Model I (about 100 Ci/yr at 3,800 years). The EBS release rates with EBS Release Model I decrease with time following the peak at about 3,800 years, indicating the release rates are controlled by the waste form alteration/dissolution rate; that is, as soon as Tc is mobilized from the waste form, it is released by the advective flux through the waste packages. With EBS Release Model II in which the EBS release rate of $^{99}$Tc is controlled by the diffusive flux through the perforations filled with the corrosion products in the "failed" waste packages, the release rates increase with time due to the increasing number of perforations, which determine the area available for diffusion through the waste package. Also, a buildup of the $^{99}$Tc concentration inside the waste packages, which causes greater concentration gradients across the waste container wall and thus higher diffusive flux through the waste packages, contributes to the $^{99}$Tc EBS release behavior.

The impact of the alternative EBS release models on the peak EBS release rate is greater for $^{135}$Cs as shown in Figure 4. The $^{135}$Cs peak EBS release rate with EBS Release Model II (about 0.03 Ci/yr at 10,000 years) is about two orders of magnitude lower than the peak release rate with EBS Release Model I (about 5 Ci/yr at 3,800 years). In general, differences in the $^{135}$Cs EBS release behavior between the alternative EBS release models are similar to those of $^{99}$Tc, and reasoning similar to that given above for the $^{99}$Tc EBS release behavior may be offered to explain the $^{135}$Cs EBS release behavior. As in the $^{99}$Tc case, the $^{135}$Cs EBS release rates calculated with EBS Release Model I appear to be controlled by the waste form alteration/dissolution rate and the advective flow rate through the waste packages. Although $^{99}$Tc and $^{135}$Cs have similar characteristics (i.e., the same gap and grain-boundary inventory, both soluble and mobile, and similar median solubilities), the lower inventory of $^{135}$Cs resulted in the lower EBS release rates compared to the $^{99}$Tc release rates. For example, the inventory contains 140 Ci $^{99}$Tc vs 5.1 Ci $^{135}$Cs per spent fuel waste package assuming the 30-year old spent fuel and the waste package characteristics in Table 1.

More significant impacts of the alternative EBS release models were seen for the radionuclides with a low solubility (or solubility-limited radionuclides). This is shown in Figure 5 for the EBS release behavior of $^{237}$Np which has a median solubility of $1.4 \times 10^{-4}$ M and a half-life of $2.14 \times 10^6$ years. With both EBS release models, the $^{237}$Np release rates increase with time, and this is caused by the increase in the number of "failed" waste packages with time (plus the increase in the number of perforations in a "failed" waste package with the second model). This indicates that, because of the low solubility of Np, its concentration at the waste form is maintained at its solubility limit (the median solubility limit in this analysis) even though the amount of $^{237}$Np that has been mobilized from the

waste form increases with time. As shown in Figure 5, with EBS Release Model II, the peak EBS release rate of $^{237}Np$ (about 0.003 Ci/yr at 10,000 years) is almost three orders of magnitude lower than the peak release rate with EBS Release Model I (about 2 Ci/yr at 10,000 years).

## SUMMARY AND CONCLUSION

Simulations were conducted to analyze the sensitivity of the EBS release rate to alternative conceptual models of the advective release from waste packages under dripping fractures. The sensitivity study was performed for the case with 83 MTU/acre thermal loading, no backfill and high infiltration. The first conceptual model, identified as EBS Release Model I or "Drip-on-Waste-Form" model, assumed that, once a waste package fails by pitting corrosion (i.e., having at least one pit penetration) and if the waste package is under dripping fractures, the dripping water directly contacts the waste form inside the waste package, and radionuclides are released from the waste package and EBS by advection. The second conceptual model, called EBS Release Model II or "Drip-on-Waste-Package" model, assumed that the perforations (or pit penetrations) in the "failed" waste package are filled with fine, gel-like porous corrosion products, and the dripping water is diverted around the waste package, thus preventing the dripping water from directly contacting the waste form. In the second model, it was assumed that radionuclides are transported through the perforations (filled with corrosion products) by diffusion, and, once outside the waste package, the radionuclides are released from the EBS by advection. The second model incorporates more realism into the EBS release calculations because it assumes that the advective release rate from a "failed" waste container depends on the number of pit penetrations. For the "failed" waste packages that were not subjected to dripping water, radionuclides were released by diffusion only. Also incorporated into the simulations were the abstractions (in simple functional forms) of the various process-level modeling results such as the near-field conditions (temperature, relative humidity and liquid saturation), waste package failure and degradation, the drift-scale hydrology (matrix and fracture flows), etc.

As expected, EBS Release Model I (or Drip-on-Waste-Form Model) yielded higher EBS release rates than EBS Release Model II for all of the radionuclides. Using EBS Release Model II (or Drip-on-Waste-Package), all radionuclides had their peak EBS release rates much lower (from at least one to several orders of magnitude) than using EBS Release Model I. The impacts of the alternative EBS advective release models were greater for the radionuclides with a low solubility (or solubility-limited radionuclides) than for those which are soluble and mobile in water (or alteration/dissolution-limited radionuclides). For example, the peak EBS release rate for $^{99}Tc$ with EBS Release Model II was an order of magnitude lower than the peak release rate with the other (conservative) model (EBS Release Model I). The peak EBS release rate for $^{237}Np$ (solubility-limited) with EBS Release Model II was about three orders of magnitude lower than the peak release rate with the other model (EBS Release Model I).

The analyses indicated that the EBS release model by advection through "failed" waste package (EBS Release Model I or Drip-on-Waste-Form) may be too conservative in evaluating the EBS performance. One major implication from this sensitivity study is that a "failed" waste package container with multiple perforations may still be able to perform as an

important barrier to radionuclide release.

## ACKNOWLEDGMENTS

This work was funded by the US DOE Yucca Mountain Site Characterization Office under Contract #DE-AC01-91RW00134 to TRW Environmental Safety Systems, Inc.

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Table 1 Waste package parameters used in the RIP simulations

| Waste Package Characteristics | Spent Nuclear Fuel (SF) | Defense High-Level Waste (DHLW) Glass |
|---|---|---|
| Number of Waste Packages | 6468 | 3829 |
| Spent Fuel Burnup (MWd/MTHM)[1)] | 39,651 (PWR)<br>31,186 (BWR)<br>36,666 (combined) | N/A |
| Mass Waste per Waste Package (MTHM/pkg)[2)] | 9.74 MTHM/pkg<br>(PWR and BWR combined) | 1.828 MTHM/pkg |

1) MWd/MTHM = megawatt days per metric tons of heavy metal
2) Total MTHM in the repository/number of waste packages

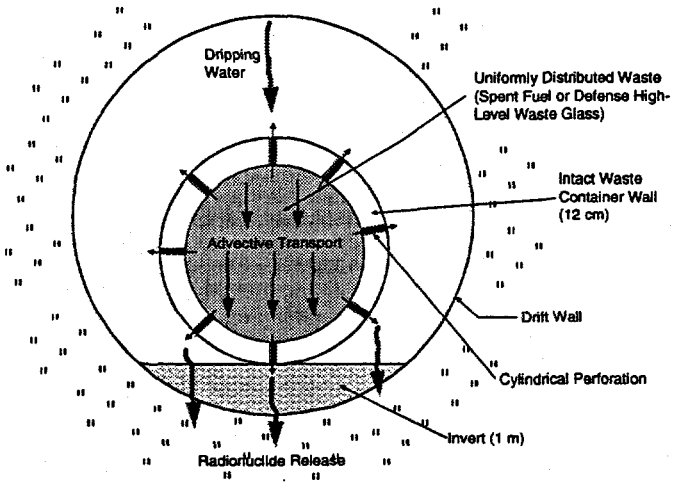


Figure 1 Schematic of the EBS advective release model I (Drip-on-Waste-Form Model) in which the dripping water directly contacts the waste form, and radionuclides are released by advection through the waste package.

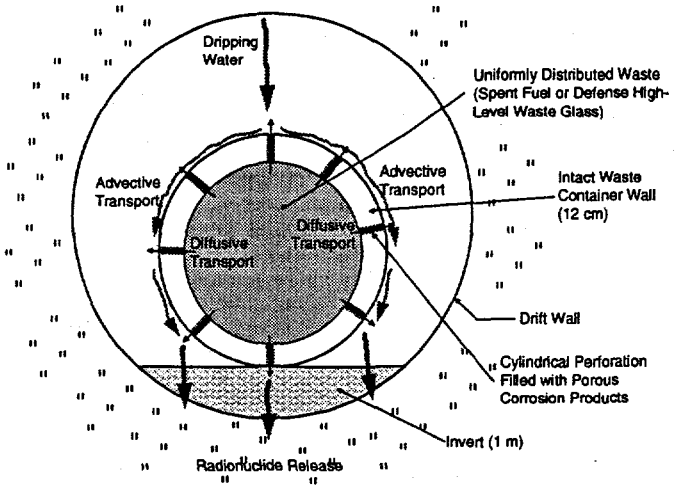


Figure 2 Schematic of the EBS advective release model II (Drip-on-Waste-Package Model) in which the dripping water is diverted around the waste package, and radionuclides are transported by diffusion through the perforations in the waste container, then are released by advection.

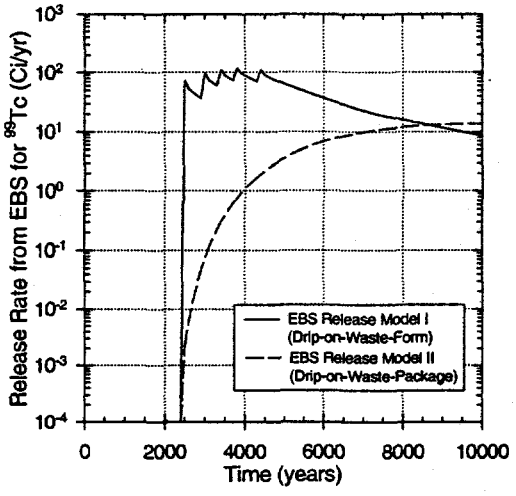


Figure 3 Sensitivity of the EBS release rate for $^{99}$Tc to the alternative EBS advective release models.

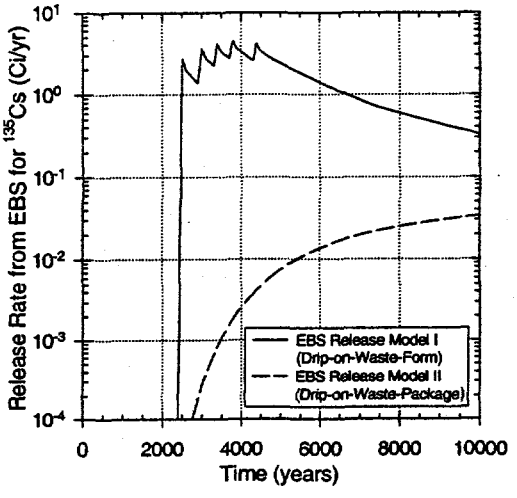


Figure 4 Sensitivity of the EBS release rate for $^{135}$Cs to the alternative EBS advective release models.

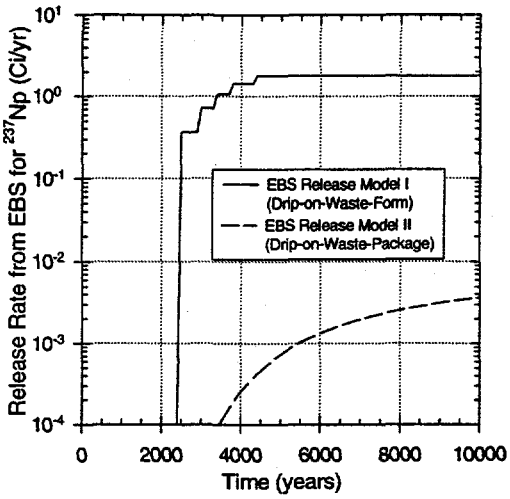


Figure 5 Sensitivity of the EBS release rate for $^{237}$Np to the alternative EBS advective release models.

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