LBL-31069
UC-814

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EARTH SCIENCES DIVISION

Preliminary Calculations of Release Rates of Tc-99, I-129, and Np-237 from Spent Fuel in a Potential Repository in Tuff

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June 1990

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Prepared for the U.S. Department of Energy under Contract Number DE-AC03-76SF00098

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This work was supported by the Director, Office of Civilian Radioactive Waste Management, Yucca Mountain Site Characterization Office, of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

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1. INTRODUCTION

This report presents preliminary calculations of time-dependent release rates of selected radionuclides from the engineered barrier system in a potential high-level waste repository in unsaturated tuff, representative of a potential repository at Yucca Mountain in southern Nevada. These results are intended for use as preliminary source terms for calculating total system performance. The radionuclides specified for preliminary release-rate calculations are Tc-99, I-129, Cs-135, and Np-237 for ground-water pathways and C-14 for gaseous release. Releases via liquid pathways are the subject of this report; UCB/LBL will prepare a separate report on C-14 gaseous releases [Light *et al.* 1990].

The USDOE has requested performance-assessment Working Group 2 to prepare these preliminary data on radionuclide release rates. This report is a joint effort of three organizations: Lawrence Livermore National Laboratory (LLNL) supports the DOE Yucca Mountain Project in the development, demonstration, and possible licensing of the Yucca Mountain potential repository site in southern Nevada; Pacific Northwest Laboratory (PNL) and the University of California/Lawrence Berkeley Laboratory (UCB/LBL) support the DOE Office of Civilian Radioactive Waste Management in its oversight role. Each of these laboratories has issued or is preparing separate reports on release rates from waste packages in tuff [Apted *et al.* 1989; Apted and Engel, 1990; O'Connell *et al.* 1989; O'Connell 1990; Pigford and Lee 1989; Sadeghi *et al.* 1990a,c]. Working Group 2 coordinates the work of the DOE groups examining the performance of the engineered barrier system. Working Group 2 agreed that for the purpose of this report they would select one set of parameters and do one set of calculations of release rates, with different water-contact modes being examined by different laboratories. LLNL made the calculations for the wet-drip water-contact modes, PNL made the calculations for the moist-continuous water-contact mode, and UCB/LBL performed independent checks of calculations of both modes and coordinated the preparation of this report.

These preliminary results are intended for use by members of performance assessment Working Group 1 for a preliminary assessment of total system performance. Working Group 1 coordinates DOE evaluations of the performance of the total mined geologic disposal system.

The calculations reported here focus on release rates from individual waste packages and from a repository-average waste package, averaged over a distribution of failure times of waste-package containers. The calculations are limited to releases from spent fuel; releases from borosilicate glass defense waste are not considered here. The release-rate results are presented as a function of time, for times up to 10,000 years after repository closure for the wet-drip contact modes and for 100,000 years and longer after closure for the moist-continuous mode. Also included, for use by Working Group 2 and others, are time-dependent fractional release rates and cumulative releases, based on the 1000-year inventories of these radionuclides.

These release results are not to be interpreted as expected values. In many instances the concepts, scenarios, models, and parameters used for the calculations are based on necessarily simplifying assumptions. In doing so, conservatism is intended to accompany simplification. Therefore, these predicted release rates are expected to be much more conservative than the releases that can be expected from more realistic assessments. Where these calculated conservative and bounding release rates are high enough to indicate concern with regard to meeting design and regulatory requirements, priority is to be given to obtaining

more realistic data, particularly chemical data and time-dependent ground-water flow regimes near waste packages, as well as exploring more realistic and more detailed models for release rates.

In Section 2 we describe the modes, or scenarios, by which water is postulated to contact the spent fuel and possible pathways by which dissolved radionuclides can be released to the surrounding intact rock, and we describe calculational models for each of these water-contact modes. In Section 3 we present and discuss the parameters adopted for calculating release rates from these models. In Section 4 we present numerical results. In Section 5 we present conclusions and recommendations. Tables of calculated results are presented as Appendices.

2. METHODS FOR CALCULATING RELEASE RATE

2.1. The Wet-Drip Water-Contact Modes

Each waste package is to be emplaced in a borehole, with an air gap between the container surface and the surrounding rock [USDOE 1988]. Water will contact the waste package only if there is some variation of the local environment away from the design condition, e.g., dripping from overhead rock or contact with moist rock [O'Connell and Drach 1986]. Possible variations in rock permeability may divert water into fractures that intersect the emplacement hole, possibly resulting in drips onto the package [O'Connell and Drach 1986]. The drips are assumed to cause local cracks in the container. We conservatively assume that all of the water dripping onto the container enters the cracks and contacts spent fuel, and we neglect protection of the uranium-dioxide fuel by the Zircaloy cladding.

The assumed penetration of dripping water into the container is assumed to result in a "bathtub" or a "flow-through" mode of water/waste contact, depending on the state of the degraded container. The "bathtub" mode occurs if breaches exist only near the top [Oversby and Wilson 1986; O'Connell and Drach 1986; Apted and Engel 1990; Pigford and Lee 1989]. Such breaches may develop if water does indeed drip onto the top of the container. Water entering the container fills the container, gradually immersing all the fuel. After filling, contaminated water from the container overflows into the surrounding rock as new water enters the container. We neglect evaporation that will occur because the container is hotter than the surrounding rock. We assume that the water within the container is well mixed. We conservatively neglect isolation of the UO_2 fuel by the Zircaloy cladding, but we assume that the general geometrical arrangement of the spent fuel within the container persists.

The rapidly releasable fractions of Tc, Cs, I and some other elements within the fuel-cladding gap, gas plenum, and readily accessible grain boundaries are assumed to all dissolve in the water by the time the container fills and overflow begins. These and other elements contained within the UO_2 matrix are released to react with container water at a rate limited by the rate of chemical alteration of the UO_2. Tc, Cs, and I from the UO_2 matrix are assumed to dissolve when released. Release of neptunium is expected to be limited by its solubility, so the release rate of Np-237 from the container is proportional to the volumetric flow rate of water into and out of the breached container.

The flow-through mode [O'Connell *et al.* 1989] is assumed to occur if there are breaches near the top and bottom of the container, or scattered throughout the original container surface. Water enters and wets a

fraction of the spent fuel (see below), moving downward as a surface film and exiting at the bottom. As the water moves it dissolves the rapid-release fraction, as well as the elements released by chemical alteration at the surface of the wetted fuel matrix. Neptunium dissolves and is released to the rock at a rate limited by its solubility. We assume that the matrix alteration rate, per unit surface area of UO_2, and the neptunium solubility are the same as in the bathtub immersed case.

The fraction of fuel wetted in the flow-through mode is a significant unknown. The residence time of water depends here on the mechanisms of wetting and drainage. It is estimated from a mass balance of the water-film volume and water influx rate. We assume that a specified fraction of fuel remains continually wetted, whether due to surface roughness or water replenishment. The resulting residence time is far less than the fill-time estimated for the wet-drip contact mode.

2.2. The Moist-Continuous Water-Contact Mode

The wet-drip modes assume that the air-filled annulus surrounding a waste package precludes pathways for diffusive release of dissolved species. However, there can exist pathways for release by liquid diffusion if a waste package contacts the surrounding rock by physical displacement, if the annulus becomes filled with sediments and rubble, or if the emplacement horizon becomes water saturated [Apted and Engel 1990; Pigford and Lee 1989]. For this report we assume rubble in the annulus, at the same moisture content as the surrounding rock.

We assume that water has filled a failed container, and we neglect mass-transfer resistance from Zircaloy fuel cladding and from the residual container material and corrosion products. We have applied the analytical solutions [Chambré *et al.* 1982 and 1985] for time-dependent mass transfer of dissolved species through porous rock to predict the time-dependent diffusive-convective release rates of radionuclides from the waste into the surrounding rock [Apted *et al.* 1989; Sadeghi *et al.* 1990c].

For the expected ground-water velocities, and for the diffusion coefficients assumed here for intact rock, mass-transfer is predicted to occur predominantly by molecular diffusion in pore liquid in the rock matrix [Pigford and Chambré 1988]. Under these conditions the release rate is finite at zero velocity and is insensitive to any but very large increases in water velocity.

We conservatively assume that all moisture in the intact rock is in interconnected water-filled pores, with a pore-liquid diffusion coefficient given by that for a liquid continuum. When calculating the time-dependent mass-transfer rate through an unsaturated rubble bed into surrounding rock, we adopt an effective diffusion coefficient several orders of magnitude lower than that of the intact rock, to account for limited contact area between rubble particles [Conca 1990; Sadeghi *et al.* 1990b].

As a result of the low effective diffusion coefficient for unsaturated rubble, with air in void spaces not occupied by rubble, the predicted mass transfer rates will be far below what could occur if the repository were saturated or if the degraded waste container were in close contact with consolidated or intact rock. Our analyses will be restricted to the unsaturated environment, so this particular scenario is more aptly termed the "moist-continuous scenario," and is so referred to herein.

To calculate the time-dependent release rate for solubility-limited Np-237, we assume a constant saturation concentration of neptunium in moisture at the waste-package surface. To calculate the time-dependent release of readily soluble species in the fuel-cladding gap, plenum, and accessible grain boundaries of spent fuel, we conservatively assume that this readily soluble inventory of I-129 and Cs-135 is released instantaneously into water that enters after the container fails. We assume that the contained water volume equals the initial void volume within the waste container. To calculate the alteration-controlled release of soluble Tc-99, I-129, and Cs-135 from the fuel matrix, we assume a constant alteration rate, as in the wet-drip contact modes, and we assume that these fission products dissolve congruently with the alteration of the fuel matrix.

3. PARAMETERS ADOPTED FOR CALCULATING RELEASE RATES

3.1 Time-Distribution of Container Failure

We adopt a probabilistic distribution of container failure as a function of time since emplacement, to yield repository-average release rates from a waste package as a function of time since emplacement. The distribution of container failures is based on the assumption that a waste container cannot fail until the rock immediately surrounding the container has cooled to the condensing temperature, so that liquid water can exist on the container surface. It is assumed that after the adjacent rock reaches the condensing temperature the container can be exposed to drips of ground water onto the container surface. The container is conservatively assumed to fail at that time, for the moist-continuous calculations, and 300 years later, for the wet-drip calculations. For this purpose we have adopted predictions [Altenhofen and Eslinger 1990] of the cumulative number of containers reaching these locally corrosive conditions as a function of time after emplacement. The failure distribution is shown in Figure 3.1.

The assumed time distribution of container failure is only partly mechanistic in terms of failure processes. It is adopted here to illustrate the importance, for some radionuclides, of incorporating a time-distribution of container failure in source-term analysis.

3.2 Waste Package Configuration

The dimensions and fuel loading of the reference waste package used in these calculations are given in Table 1.

Table 1. Dimensions and Fuel Loading of Reference Waste Package

Waste Package Height	4.6 m
Waste Package Outside Diameter	0.71 m
Waste Package Overall Volume	1.73 m^3
Waste Package Internal Void Volume	1.22 m^3
Waste Package Content	2.1 Mg U in original fuel (2.1 MTHM)
Surface area of intact fuel (without cladding)	140 m^2
Air gap between waste container and emplacement hole	3 cm

3.3. Water Infiltration Rate in Partially Saturated Tuff

Hydrological properties used in these calculations are given in Table 2.

Table 2. Hydrological Properties for Calculating Release Rates

Far-field average Darcy velocity of ground water	0.5 mm/year
Water drip rate onto waste package	1 liter/year
Assumed fraction of fuel rods wetted by ground water in the wet-drip flow-through model	0.20
Travel time for water in waste package, wet-drip flow-through model	50 years
Porosity of tuff surrounding a waste package, and porosity of pieces of tuff rubble	0.16
Water saturation of intact tuff surrounding a waste package, water saturation of pieces of tuff rubble	0.86
Void fraction in tuff rubble bed (volume of air surrounding rubble pieces/total volume of rubble bed)	0.30

Temperature does not appear in the present calculations, except as an indication of when liquid water may be excluded or present, as discussed in Section 3.1. The far-field average flux (Darcy velocity) of ground water is likely to have local variations. Many waste packages may remain essentially dry, i.e., no water influx and no substantial continuous pathway for diffusion. Some waste packages may have advective water contact modes or a diffusive water contact mode, for which further properties of the local hydrologic conditions must be established.

The far-field averaged flux adopted for this study is 0.5 mm/year. This is expected to be an upper bound on the average water flux for anticipated conditions [USDOE 1988]. Variations from this value should also be evaluated in future studies.

As a performance allocation, the DOE has assumed that more than 90 percent of the boreholes will remain "dry," i.e., no weep of ground water on the borehole wall, no dripping, and no substantial continuous liquid pathway for diffusion [USDOE 1988, Secs. 8.3.4.2, 8.3.5.9 and 8.3.5.10]. Possible gaseous releases during the dry period are considered in a separate report on $^{14}CO_2$ release. LLNL expects such dry conditions because the 0.5 mm/y flux can be carried by porous flow in the unsaturated rock matrix, with no net flow in fractures that could lead to dripping. Also, the gap or partial gap at the drift ceiling would similarly serve to keep water in the rock matrix, diverting flow away from the drift and borehole. A potential continuous standing pathway, e.g., porous rock and surface films of water, would be broken by the air gap between container and borehole wall, unless some rock movements occur or unless the gap becomes filled with exfoliated porous material and rubble. Metal surfaces, cracked container, and internal geometry would also provide barriers to a diffusive water pathway. Nitao [1988] has pointed out that, under partially saturated hydrological conditions, water would be preferentially absorbed by tuff with its small-radius grain and pore sizes. To maintain a water film through container cracks and over spent fuel elements, development of small-radius surface texture, i.e., high surface roughness or low-angle contacts, might be necessary.

Some variations away from these usual conditions are to be expected at some of the waste packages. We do not yet have a detailed or bounding mechanism-based explanation for such variations. We assume, consistent with the *Site Characterization Plan* [USDOE 1988] performance allocation, that up to 10 percent of the waste packages could experience dripping or diffusive pathways.

For dripping water at some boreholes, we adopt a value of water flux by reference to the amount of water passing through an undisturbed cross-sectional area above the borehole. Disturbances from the surrounding or variations in local hydrologic properties would in some cases divert water away from the borehole, but perhaps in some cases divert water toward the borehole. We assume a larger "catchment area" of radius two times the borehole radius. This gives a water influx of 1.0 liter/year into a breached container.

3.4. Diffusion Coefficient in Partially Saturated Tuff

The diffusion coefficient for radionuclides in a water continuum, 1×10^{-5} cm^2/s, has been previously used in release calculations for waste packages emplaced in tuff [Apted *et al.* 1989; Pigford and Lee 1989]. When used for calculating diffusion through porous media, this value conservatively ignores tortuosity reduction of the effective diffusion coefficient and of the release rate.

For transport through an unconsolidated rubble bed of partially saturated porous media, there are additional tortuosity effects. Under the partial-saturation conditions expected at the emplacement horizon at Yucca Mountain, no water films are expected on the surfaces of rubble pieces. Transport is constrained to diffusion through interconnected water-filled pores within rubble pieces, through areas of contact between adjacent rubble pieces, through contact areas between rubble pieces and intact rock, and through contact areas between rubble pieces and the surface of the degraded waste container. Diffusion through pore liquid in contacting rubble pieces and through contact areas has received preliminary examination by experiments and by theoretical analysis for diffusional transport through beds of tuff gravel [Conca 1990; Sadeghi *et al.* 1990b]. Both studies show that for the degree of partial saturation assumed for the Yucca Mountain site, the effective diffusion coefficient through gravel-sized rubble pieces is likely to be many orders of magnitude below that of intact rock. A thousand-fold reduction below that of intact rock is adopted for this study.

The diffusion coefficients quoted here are used in a Fick's-Law expression to calculate the diffusive transport per unit cross-sectional area of pore liquid, the proper formulation for calculating transient releases. A consistent value of the porosity must be used in the transport boundary condition, as discussed in the next section.

3.5. Effective Porosity in Partially Saturated Rock and Rubble

In calculating the Fick's-law diffusive flux (g/cm^2-s) through pore liquid in unsaturated intact rock, the effective porosity that appears in the flux boundary condition at an interface is the product of the rock-matrix porosity ϵ_r and the saturation fraction Ψ, i.e., the fraction of the pores that are water filled. This assumes isotropic porosity and continuous diffusive pathways through water-filled pores. The $\epsilon_r \Psi$ product is defined as the liquid transport porosity ϵ_t for intact rock and is used wherever the porosity appears in equations derived for mass transport through an equivalent saturated porous medium. It neglects the fracture porosity of intact rock, only a small correction for unsaturated tuff.

To calculate the sorption retardation coefficient for transport in unsaturated porous rock, assuming local chemical equilibrium between a species in the pore liquid and that same species sorbed in the rock, the retardation coefficient K is given by

$$K = 1 + \frac{1-\epsilon_r}{\epsilon_r \Psi} K_d \tag{1}$$

where K_d is the dimensionless distribution coefficient (mass per unit volume of rock solid divided by mass per unit volume of pore liquid)

ϵ_r is the overall porosity of the rock matrix

Ψ is the fractional saturation of the rock matrix.

Values of the distribution coefficient and calculated values of the retardation coefficient are listed in Table 3, in Section 3.6.

For a gravel bed of unsaturated rock pieces, we define ϵ_b as the rubble-bed porosity, i.e., the volume of air between rubble pieces divided by the total bed volume. Here the effective porosity ϵ_t that appears in the flux boundary condition at an interface, and that appears explicitly in the transport equations for an equivalent saturated porous medium, is approximated by

$$\epsilon_t = (1-\epsilon_b)\epsilon_r \Psi \tag{2}$$

For the porosity and saturation values in Table 2, the effective porosity for transport is 0.096.

The retardation coefficient in rubble pieces is still given by Equation (1), assuming that the rubble consists of small pieces of intact rock, of the same porosity and saturation as intact rock.

3.6 Radionuclide Data

The radionuclides selected for release-rate calculations are Tc-99, I-129, Cs-135, and Np-237. Radionuclide data used in these calculations are shown in Table 3. These data include solubilities (where appropriate), sorption and retardation coefficients on tuff, and inventories in spent fuel. The sorption coefficients are from a written communication from Eggert, Los Alamos National Laboratory, and are similar to previously published values [USDOE 1986; USDOE 1988]. The inventory data are taken from USDOE [1987, Vol. 1, Tables 2.4.2 and 2.4.6]. The solubility for Np-237 is from Bruton and Shaw [1988] and is discussed in Section 3.7.2.

Table 3. Radionuclide Data For Calculation Of Release From Spent Fuel

Nuclide	1000-yr Inventory (Ci/package)	Solubility (g/m^3)	Distribution Coefficient*	Retardation Coefficient
Tc-99	26.0	——	0.1	1.6
I-129	6.30×10^{-2}	——	0	1
Cs-135	6.90×10^{-1}	——	100	610
Np-237	2.00	3.0×10^{-4}	5	32

*Concentration in solid/concentration in liquid

Some of these radionuclides are found in different forms and locations in spent fuel. Mobile Tc-99, I-129 and Cs-135 occur in both the UO_2 matrix and in the fuel-cladding gap, gas plenum, and accessible grain

boundaries. In this document fission products in those regions other than the UO_2 matrix are referred to as the "gap" fission products. The proportion of radionuclides in the gap can vary widely because of varying conditions during reactor operations. In this report we assume that 2 percent of the total inventory of Tc-99, I-129, and Cs-135 is in the gap, a reasonable median value for light-water-reactor fuels [Apted *et al.*, 1989]. Furthermore, the gap release calculated in this study can be directly scaled if some alternate value for gap inventory is proposed. The gap release rate is proportional to gap inventory.

The selection of the four radionuclides in Table 3 is based on several considerations of the important radionuclides for far-field transport and overall system performance. Because 1000 years of water travel time is assumed for total system assessment, no short-lived radionuclides (e.g., Sr-90, Cs-137) need be considered. The half-lives for the selected radionuclides range from 2.15 $\times 10^5$ to 1.59 $\times 10^7$ years. Fission products (Tc-99, I-129, Cs-135) and actinides (Np-237) are represented. Solubility-limited (Np-237) and reaction-rate-limited (Tc-99, I-129, Cs-135) nuclides in the matrix are included. Tc-99, I-129 and Cs-135 are also contained in the gap phase. Finally, there is a range in sorption properties of these selected radionuclides, from non-sorbing I-129, weakly sorbing Tc-99 and Np-237, to strongly sorbing Cs-135.

3.7 Chemical Controls on Radionuclide Release from UO_2

The release of radionuclides contained within the UO_2 matrix of spent fuel will be affected by the same chemical parameters of these radionuclides, regardless of the release pathway (wet-drip or moist-continuous). Upon contact with water, radionuclide releases may be constrained by the solubility of the UO_2 matrix, the solubility of the radionuclide-bearing precipitate, or the rate of physicochemical alteration of the UO_2 matrix. Which of these processes is, in fact, limiting will depend on factors such as the chemistry of the given nuclide, the abundance of the nuclide within the UO_2 matrix, the chemical composition of the contacting water, the temperature, and, in particular, the redox potential.

3.7.1 Composition of Water

The anticipated composition of vadose water at the Yucca Mountain site is currently represented by J-13 well water. The reported *p*H of this water is 6.9 and the total dissolved carbonate concentration is 2.3 $\times 10^{-3}$ M [USDOE 1988, Chapter 4, Tables 4-6, 4-7] at 25 C. The vadose water and atmosphere in the emplacement horizon at the candidate Yucca Mountain site are projected to be mildly oxidizing [USDOE 1988, p. 7-9]. This is due to the exchange of emplacement-level air with the external environment, with possible contributions from radiolysis. The ambient atmospheric O_2 fugacity would be $10^{-0.7}$ bar at 25 C. Based on evaluation of test data, Wilson and Bruton [1989] postulate that buffering by the spent fuel or by metallic portions of the engineered barrier system may locally impose a low O_2 fugacity at the UO_2 surface. Grambow [1989] has also suggested that the oxidative reaction of spent-fuel matrix may locally buffer the O_2 fugacity to reducing values (estimated O_2 fugacity of 10^{-60} bar at 25 C).

Within this large range in the possible redox conditions, the UO_2 matrix is expected to be unstable with respect to higher-valence uranium compounds. For example, oxidation to U_3O_8, which is not isostructural with UO_2 or U_3O_7, or with lower-valence uranium oxides, can expose matrix constituents to reaction with water. Therefore, if there is sufficient dissolved oxygen to form U_3O_8 within the failed container, matrix

solubility is not likely to control the release of other matrix constituents for the Yucca Mountain site. In this case, the release of some radionuclides (e.g., most actinides) as solute in water is expected to be limited by the formation of new solubility-limiting solids. However, for other nuclides (e.g., Se-79, Tc-99, I-129, Cs-135, and Cm-245) of sufficient solubility and/or sufficiently low concentration in the waste matrix, the alteration rate of the UO_2 matrix may limit their release as solute to the ground water. Alteration-limited releases of Tc-99, I-129, and Cs-135 are calculated for this study.

3.7.2 Solubility Data

The solubilities of radioelements released from spent fuel under expected conditions at Yucca Mountain have been estimated through laboratory and modelling studies [Bruton and Shaw 1988; Wilson and Bruton 1989]. The precise solubility values to use in release modelling are uncertain for several reasons, however. There are inherent uncertainties that arise from measurement uncertainty and that, in turn, are passed along into thermochemical databases. There is uncertainty in the chemical environment of the near field and its evolution over time, particularly during the thermal period. Fortunately, the chemical composition of water is expected to change only slowly after liquid-saturation conditions are established in the near field. There is uncertainty in determining which solubility-limiting solid will form, its degree of crystallinity, and the effect of solid-solution substitution of other chemical components. Model projections for solubilities are particularly problematical. This is because the model is limited by the comprehensiveness of its database. Bruton [1988], for example, has noted that the thermodynamic databases for most radioelements are skewed toward oxide, hydroxide and halide compounds and aqueous species. In many cases, however, phosphates, sulfates, silicates, or alumino-silicates may be the actual solubility-limiting compounds that form. Despite these caveats and present limitations, a solubility value for Np (assuming a NpO_2 solid) of 3.0×10^{-4} g/m^3 is used based on data from Wilson and Bruton [1989].

3.7.3 Alteration-Rate Data

Wilson and Bruton [1989] evaluated the alteration rate of irradiated UO_2 from a sample of light-water-reactor spent fuel under conditions simulating a repository located in tuff. Their analysis is based on the observed release rate of soluble radionuclides (Tc-99, Cs-137, I-129 and Sr-90) into J-13 water, for exposure times up to 1050 days and for five cycles of exposure to new batches of J-13 water per fuel sample [Wilson 1987a,b]. Their estimates for UO_2 alteration rate range between 1.2×10^{-3} and 10^{-4} parts released per year for these short-term tests. These values are derived from the mass inventory and from measured dissolved concentrations of soluble nuclides.

Alteration of UO_2 in contact with water begins with the oxidation of UO_2 to higher-valence isostructural oxides, such as U_4O_9 and U_3O_7 . The dissolution reaction of U_3O_7 with water and further oxidation lead to new solid phases, e.g., U_3O_8, that are not isostructural with UO_2. Thus, we speak of alteration by oxidative dissolution, even though there may be no net dissolution, i.e., no increase in concentration of uranium in the liquid phase. Such alteration by oxidation dissolution is referred to as alteration or dissolution in this report.

In a recent review of the published literature on oxidative dissolution of UO_2 Grambow [1989] provides a

useful summary of the chemical factors that affect the alteration of UO_2. Grambow addresses three principal chemical factors, pH, dissolved carbonate concentration, and redox potential (oxygen fugacity). The effects of these factors are summarized as follows:

o pH - There appears to be an inverse, linear dependence between pH and the log of the UO_2 reaction rate, especially at pH>4.

o Dissolved carbonate - Available studies support a direct proportionality between total carbonate and dissolution rate of UO_2, especially at concentrations below 10^{-3} M. An alternate interpretation is that there is no direct effect of carbonate concentration on UO_2 reaction rate, and that the observed effects are attributable to the increase in uranium solubility with increasing concentration of carbonate as a complexing anion.

o Redox potential - The rate of UO_2 dissolution is sensitively related to the redox potential, although exact functional dependence varies with respect to pH and carbonate concentration because of changes in rate-limiting mechanisms [Grambow, 1989]. Both linear and square-root dependence of UO_2 dissolution rate on the redox potential have been proposed; for low oxygen fugacities ($< 10^{-0.7}$ bar), a linear dependence seems to provide the best fit to available data, according to Grambow [1989].

For the anticipated J-13 well water composition and an oxygen fugacity of $10^{-0.7}$ bar, the functional relationships drawn by Grambow [1989] indicate a range in values between $10^{-0.1}$ and $10^{-3.0}$ g/m^2-day for the rate of dissolution alteration of UO_2. A maximum UO_2 surface area of 140 m^2 is calculated based on complete exposure of the fuel rods contained in one standard waste package (cf. Section 3.1). For a uranium inventory of 2.1 MTHM (2.1 Mg of original uranium) per waste package, a range in fractional dissolution rates between 2.3×10^{-3} and 2.3×10^{-5} parts per year is estimated. Based on the studies of Wilson and Bruton [1989] and Grambow [1989], a high value of 10^{-3} parts per year is selected for the fractional alteration rate of UO_2 for the moist-continuous-mode calculations. For the wet-drip-mode calculations, the value of 1.2×10^{-3} parts per year is selected, based on the highest long-term rate in the experiments summarized by Wilson and Bruton [1989]. For the purpose of this report, we assume a constant mass rate of alteration, so alteration is complete after 1000 years of exposure to water.

An unaccounted factor in this analysis is the effect of near-field boiling on the composition of vadose water. Calculations by Arthur and Criscenti [1989] show that boiling under open-system conditions reduces dissolved carbonate (exsolving of CO_2 gas, as well as dissolved O_2), with a corresponding increase in pH. This open-system boiling also increases the concentration of other aqueous species and the overall solubility. Also, the concentration of other strong complexants, such as phosphates, will increase. These complications increase the uncertainty for the solubility values and reaction rates. The reaction rate of UO_2 would be greatly reduced in such modified water compared to the assumed oxygenated J-13 water. Because every unit increase in pH leads to a reduction in UO_2 alteration/dissolution rate by a factor of 10, the effect of boiling may be a key process that affects source-term calculations. Further modeling and laboratory validation of the effect of open-system boiling are needed.

4. CALCULATED RELEASE RATES

4.1 Release-Rate Calculations

The authors of this report represent three of the laboratories (LLNL, PNL, and UCB/LBL) that support DOE and the Yucca Mountain Project in its performance assessment of waste-package performance. Calculational techniques from each of these laboratories have been reviewed and have been shown to yield similar results for the same mode of water contact and for the same input parameters. Each of these laboratories has issued or is preparing separate reports of calculations of release rates from waste packages in Yucca Mountain [Apted *et al.* 1989; O'Connell 1990; Pigford and Lee 1989, Sadeghi *et al.* 1990a,c]. Working Group 2 agreed that for the purpose of this report there should be one calculation of release rates from parameters agreed on for this purpose. The Lawrence Livermore National Laboratory made the calculations for wet-drip releases, and the Pacific Northwest Laboratory made the calculations for moist-continuous releases. Calculations by UCB/LBL of wet-drip and moist- continuous release rates from individual waste packages, including an analysis of the effect of flow on moist-continuous release rates, are already presented elsewhere [Pigford and Lee 1989; Pigford and Chambré 1988).

4.2 Wet-Drip Release Rates

The wet-drip release rates from a single waste package are calculated for the scenarios described in Section 2.1. Releases congruent with fuel-matrix alteration are the dominant releases for Tc-99, I-129, and Cs-135. The releases for these three species for the bathtub model are shown in Figure 4.2.1. The first release occurs at about 2900 years, assuming that water first reaches and penetrates a cooled container, followed by a calculated fill time of about 1200 years, after which the container water overflows to the surrounding rock. Assuming that the amount of fuel exposed to water increases linearly as the container fills, the last of the fuel begins alteration just before overflow. The concentration in the container liquid and the release rate increase further for a thousand years. Thereafter, the release rate decreases exponentially with time because of dilution of the well-mixed container liquid with more ground water flowing in through penetrations.

The time-dependent release rates of Tc-99, I-129, and Cs-135 for the flow-through mode are shown in Figure 4.2.2. The first liquid flowing from the bottom of a failed container, at about 1750 years, carries the start of a pulse of these species from the readily soluble gap inventory. This assumes that the container is wet and breached at 1700 years and an additional 50 years to accumulate enough water droplets and films to start draining water, i.e., about 50 liters of water covering a fraction of the 140 m^2 surface areas of intact fuel. The full gap inventory is then assumed to be released over the next 50 years. For the matrix alteration-controlled release rate, it is assumed that 20 percent of the fuel surface is wet at any time, with all the fuel progressively contacted by water. Thus, the alteration-controlled release rate for the flow-through mode is only 20 percent of that for the bathtub model, but it lasts five times longer.

The matrix-component release rate of Tc-99 and I-129 is bounded by the matrix alteration rate, but it could be much lower, proportional to the amount of fuel wetted. The prompt-release rate depends on the average residence time of water droplets on the spent fuel surface and on the time for water to contact the prompt-release component on all the fuel. Both release rates are influenced by the container fill time.

The Np-237 release rate, shown in Figures 4.2.1 and 4.2.2, is controlled by neptunium solubility, together with the ground-water flux. The solubility control results in a lower dissolution rate than would occur from the matrix alteration. Therefore, as the fuel matrix is altered neptunium is released from the fuel matrix and forms a neptunium precipitate at the fuel surface. The precipitate slowly dissolves in the flowing ground water.

The single-package release rates of Figures 4.2.1 and 4.2.2 can be used with a specified time distribution of container failure to obtain a release rate for the repository-average waste package, for each water-contact mode. For this purpose the cumulative distribution function estimated from the time-temperature calculations of Altenhofen and Eslinger [1990] is approximated as a linear distribution. The repository-average time-dependent release rates for Tc-99 are shown in Figure 4.2.3 for the bathtub model and in Figure 4.2.4 for the flow-through model. As a result of the averaging, the repository-average peak release rate of Tc-99 for the bathtub model is about a third less than the peak release rate for a single package. For the flow-through model the early spike release of readily-soluble gap Tc-99 nearly disappears for the repository-average waste package, and the release rate of alteration-controlled Tc-99 is unchanged.

Release rates of soluble I-129 and Cs-135, shown in Figures 4.2.3, and 4.2.4, follow the same time-averaging pattern as Tc-99.

For Np-237, shown in Figures 4.2.3 and 4.2.4, the time averaging results in a linearly increasing release rate, followed by the same steady-state release rate as for a single waste package.

The tabulated release rates from a single waste package and from a repository-average waste package are shown in the Appendix for the bathtub and flow-through models.

4.3. Moist-Continuous Release Rates

The moist-continuous release models used in these calculations are derived from original development by Chambré *et al.* [1982] and Chambré *et al.* [1985]. Using these equations and the parameters described above, we calculate release rates and cumulative releases from the annular rubble bed into the surrounding intact rock. The results are divided into two parts. The first part describes the calculated releases from a single waste package assumed to fail 1000 years after repository closure. The second part describes the calculated releases from an engineered barrier system, presented as the cumulative release from a repository-average waste package. Release is assumed to begin when a container fails. The container failures of these waste packages are assumed to be distributed in time according to the distribution derived by Altenhofen and Eslinger [1990].

4.3.1 Release from an Individual Waste Package

The calculated release rates of nuclides from the UO_2 matrix are shown in Figures 4.3.1 - 4.3.5. Fractional release is defined as release normalized to the 1000-year inventory for that nuclide, as specified in 10 CFR Part 60 [U.S.N.R.C., 1983]. Among nuclides limited by matrix-alteration rate, Figures 4.3.1-4.3.3, the release rates of low-sorbing Tc-99 and I-129 peak at approximately 1000 years after container failure. Both of these nuclides approach, but do not reach, the maximum fractional release rate equal to the assumed fractional

alteration rate of the UO_2 matrix, i.e., 10^{-3} parts per year. The release-rate curve for non-sorbing I-129 rises somewhat faster than that for slightly sorbing Tc-99.

The peak release rate of strongly sorbing Cs-135 from the matrix, shown in Figures 4.3.1-4.3.3, is delayed for several thousand years after container failure. The peak fractional-release rate of Cs-135 occurs later and is lower than that of Tc-99 and I-129, even though all three are assumed to be released congruently with matrix alteration. The long delay in release of Cs-135 to the rock is a consequence of the low diffusion coefficient in the rubble annulus and the high sorption of cesium. This delay allows time for diffusive dispersion of the cesium front, resulting in lower peak releases than the weakly sorbing Tc-99 and I-129. The delay is not sufficient for decay of Cs-135 to affect its peak release rate to the surrounding rock. The possibility of a lower release rate of cesium because of cesium solubility has not been included in the present calculations. Nor have we considered limits to cesium mobility due to the formation of a cesium aluminosilicate.

The release rate of Np-237 contained in the UO_2 matrix, shown in Figures 4.3.1-4.3.3, is solubility-limited rather than alteration-rate limited. It rises to a peak fractional-release rate of approximately 10^{-10} per year at about 100 years after container failure.

The calculated results for release of the readily soluble gap activity (i.e., 2 percent of the inventory for Tc-99, I-129, and Cs-135) are presented in Figures 4.3.4 to 4.3.6. As expected, the release rates for low-sorbing Tc-99 and I-129 rise more quickly than for the strongly sorbing Cs-135. The release rate of Tc-99 rises to half the peak rate at about 5 years after the beginning of release. The peak releases for Tc-99 and I-129 occur within a few tens of years after container failure. The peak release for the strongly sorbed Cs-135 appears several thousand years after container failure. The peak release rate of readily soluble Cs-135 is attenuated by dispersion.

The effective diffusion coefficient of radionuclides within the annulus of crushed gravel between the waste container and the host rock is a key parameter for the moist-continuous case. It significantly affects the rate of releases of the soluble gap activity and of strongly sorbing cesium from matrix alteration. The assumed value of the rubble-bed diffusion coefficient used in these calculations is 10^{-8} cm^2/s. This is likely to be a conservative value, suggested from laboratory measurements and theoretical predictions, as described previously.

4.3.2 Release from the Engineered Barrier System

Figures 4.3.7 - 4.3.9 present the calculated cumulative releases from an average waste package in the potential repository for the moist-continuous contact mode. These cumulative releases were obtained by convoluting over time the release rate from an individual package and the probability distribution of waste-package failure. A nearly uniform rate of container failure between 300 and 2300 years after waste emplacement is assumed. This distribution is from Altenhofen and Eslinger [1990], Figure 3.1, as described earlier. We assume that ambient moist conditions attain at the waste-container surface (as calculated by Altenhofen and Eslinger) at the time of container failure. Furthermore, we assume that each waste package, once failed, releases these four radionuclides at rates, as a function of time since failure, given by Figures 4.3.1- 4.3.6. No temporal or spatial variation in waste package performance is considered. For the time scales of interest

here, up to a few hundred thousand years, the half lives of all of the nuclides considered here are long enough that no decay corrections are needed.

At about 3,000 years after waste emplacement, the entire inventories of Tc-99 and I-129 have been released to the tuff rock surrounding the engineered barrier system. Their relatively rapid releases are attributable to their relatively high solubility, their low sorption coefficients for tuff, their initial release from readily-soluble gap activity, and the assumed rapid alteration rate of UO_2. Most of the inventory of Cs-135 is calculated to be released into the intact tuff rock by about 30,000 years after waste emplacement. The fractional cumulative release of solubility-limited Np-237 is significantly lower than that of the other nuclides.

5. CONCLUSIONS, OBSERVATIONS, AND RECOMMENDATIONS

Calculated time-dependent release rates of Tc-99, I-129, Cs-135, and Np-237 have been calculated for wet-drip and moist-continuous scenarios. Subject to the parameters adopted for these calculations, these results and the foregoing discussion lead to the following conclusions, observations, and recommendations:

1. The peak single-package release rates for the wet-drip bathtub and moist-continuous contact modes are within a factor of two for Tc-99 and also for I-129. In both modes, the waste-form alteration rate sets a limit on the peak release rate.

2. The peak release rate of Cs-135 is more than an order of magnitude lower for the moist-continuous mode than for the wet-drip mode.

3. The peak release rates of Np-237 are the same for the wet-drip bathtub and flow-through contact modes. Np-237 release is delayed longer for the bathtub mode because of the time to fill the waste container.

4. The peak release rate of Np-237 for the moist-continuous mode is about the same as for the wet-drip modes, for the wet-drip flow rate assumed in these calculations. For all contact modes the fractional release rates of Np-237 are much lower than for the other nuclides considered here, because of the low solubility and high inventory of Np-237.

5. The low peak release rates for the moist-continuous mode are a consequence of the very low diffusion coefficient expected in the rubble zone between waste and rock for the unsaturated environment.

6. The rate of alteration of UO_2 by oxidative dissolution controls the release rates of Tc-99, I-129, and Cs-135 in both contact modes. There is much uncertainty in the alteration rate constant used, but the adopted value of about 10^{-3}/yr is expected to be conservative. The alteration rate per unit surface area of UO_2 and the surface area of UO_2 contacted by water are among the most important release-rate parameters for further study, both experimental and theoretical.

7. Release of Np-237 is controlled by neptunium solubility. The solubility is uncertain because of uncertainties in the chemical conditions within the waste package. The calculated release rate of Np-237 may be low enough that conservatism in calculations can sufficiently allow for

uncertainties in solubility.

8. Uncertainties in ground water flow rate have little effect on calculated peak release rates of alteration-controlled Tc-99, I-129, and Cs-135 for either water-contact mode. Uncertainties in flow introduce corresponding uncertainties in Np-237 release rates for the wet-drip contact mode, but not for the moist-continuous mode, unless very large increases in ground water flow are postulated or unless the flow becomes sufficient to saturate the rock locally, increasing the effective diffusion coefficient in the rubble zone. The assumed value of the volumetric flow rate of water that reaches neptunium saturation in the wet-drip contact mode needs validation.

9. Release rates of radionuclides for the moist-continuous scenario increase by more than an order of magnitude if a significant portion of the degraded container makes contact with intact rock or if the emplacement horizon becomes fully saturated, bypassing the otherwise high diffusion resistance of unsaturated rubble between the container and intact rock.

10. Element-specific data are needed for the effective diffusion coefficients for liquid transport in unsaturated intact tuff. Further study of the diffusion coefficient in rubble is also needed.

11. Container failures distributed over times greater than the UO_2 alteration time can reduce the repository-average release rates of Tc-99, I-129, and Cs-135. Even narrower time distributions of container failure can eliminate the single-package early release spikes for the wet-drip flow-through scenario.

12. A better means of predicting the time distribution of container failure and the size of container penetrations that cause failure is needed.

13. The present release calculations assume no resistance to flow or mass transfer from the mostly-intact failed containers and fuel cladding and from their corrosion products. More detailed models that take these resistances into account should be explored.

14. We need more-detailed models and calculations that account for the availability of oxygen and of other oxidizing species, to predict UO_2 alteration rates and solubilities better. Also to be considered are the chemical changes (e.g., pH, dissolved carbonates, ionic strength) caused by distillation and localized condensation of ground water before the water reaches waste packages.

15. The four radionuclides considered in these studies are important for calculating long-term system performance, including calculations of concentrations in contaminated water reaching the environment. In calculating the cumulative release of radionuclides to the accessible environment, Tc-99 and Np-237 are potentially more important in assessing compliance with EPA's cumulative-release limit, because of their greater inventories.

16. Also important are the release rates of Th-230, U-234, and U-238, which affect the long-term release of Ra-226 and Pb-210 to the environment. Release of C-14 via ground water is important for scenarios that can result in ground water travel times of a few thousand years or less. Other potentially important long-lived radioelements include nickel, selenium, zirconium,

niobium, palladium, plutonium, americium and curium [Oversby, 1987].

17. The calculations presented herein do not consider the effects of possible colloid formation and colloid-solute interaction on release rates. These effects should be included in studies of colloid transport.

18. Whether or not any of these water-contact modes actually occurs can make a large difference in the release rates of all of the radionuclides examined. To determine when and if either water-contact mode occurs for individual waste packages, information is needed on the time-dependent ground-water flow regimes at the local scale, on borehole wall integrity, and on variability of these properties among waste-package locations.

19. The water-contact modes considered herein assume that water can penetrate the container and contact spent-fuel waste once the container material has cooled sufficiently to condense water. However evaporation of water within a breached container, because of the hotter fuel surfaces, can still limit waste-water contact in both the wet-drip and moist-continuous modes. Studies of evaporation as affected by hydrology and heat-transfer, and studies of the local chemical environment as affected by evaporation, are needed for more realistic evaluations of release rates.

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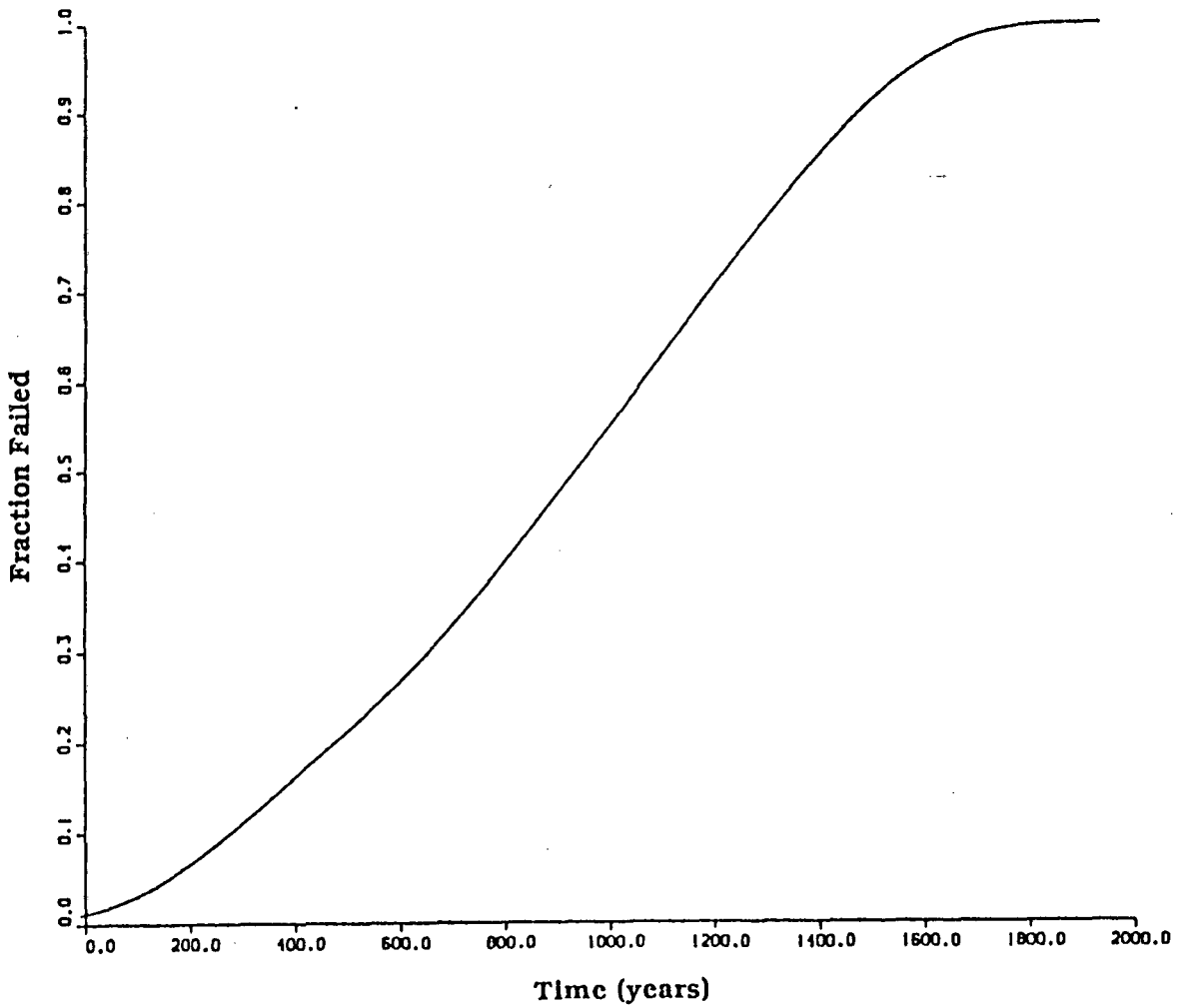

Figure 3.1 Assumed Time-Distributed Container Failure Rate

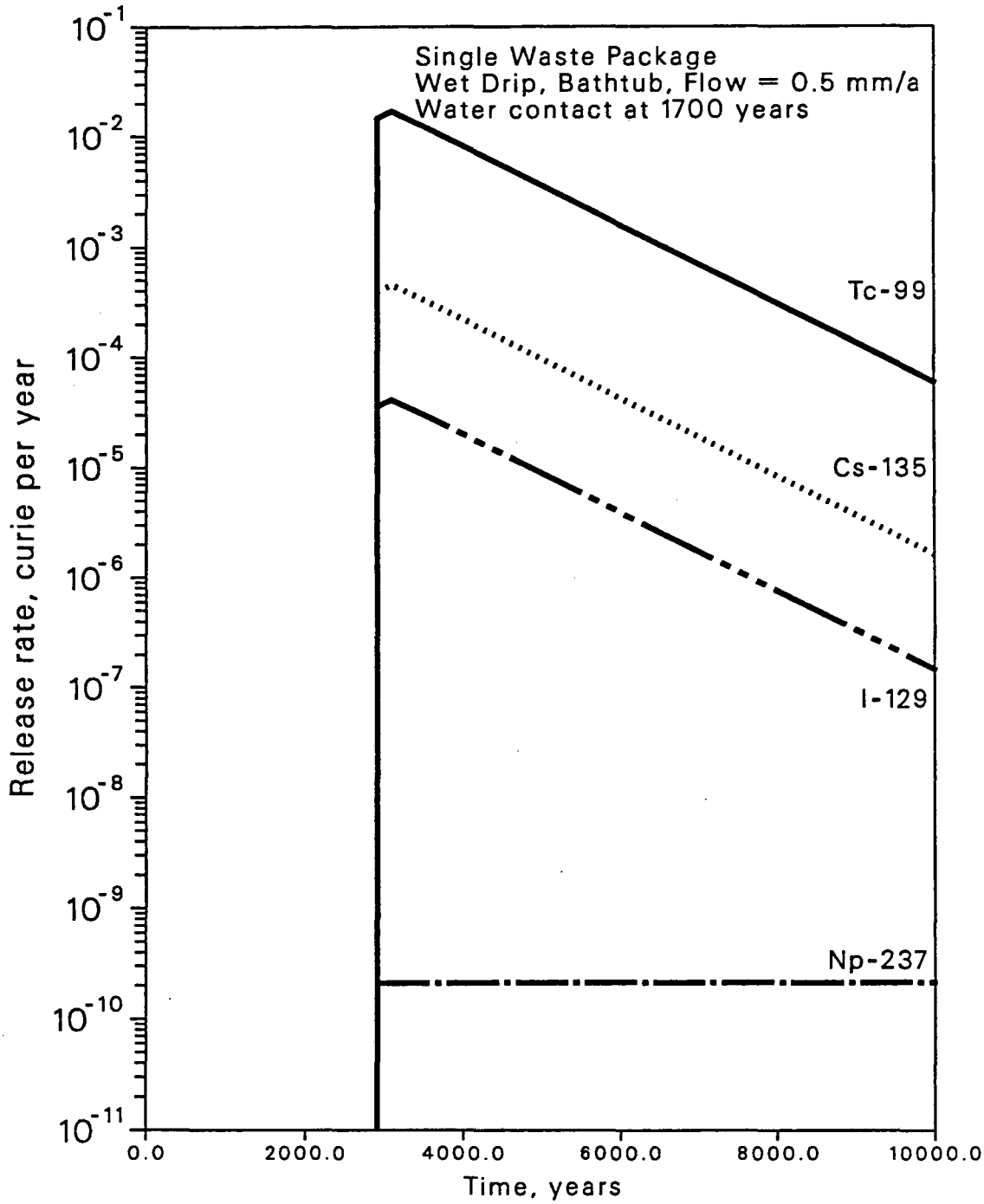

Figure 4.2.1 Release of Tc-99, I-129, Cs-135, and Np-237 in Curies/Year, for a Single Package Containing Spent Fuel, Wet-Drip Bathtub Contact Mode

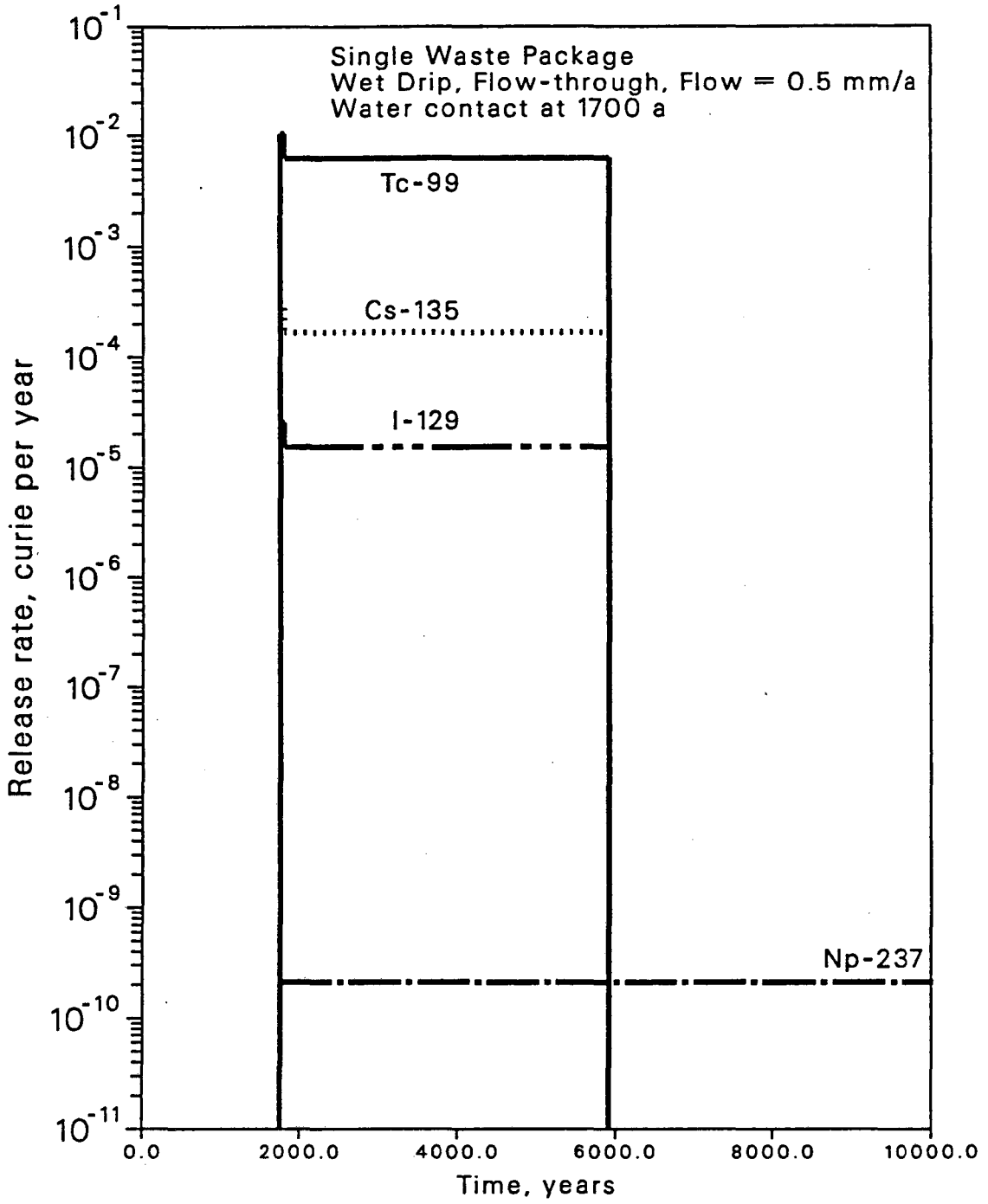

Figure 4.2.2 Release of Tc-99, I-129, Cs-135, and Np-237 in Curies/Year, for a Single Package Containing Spent Fuel, Wet-Drip Flow-Through Contact Mode

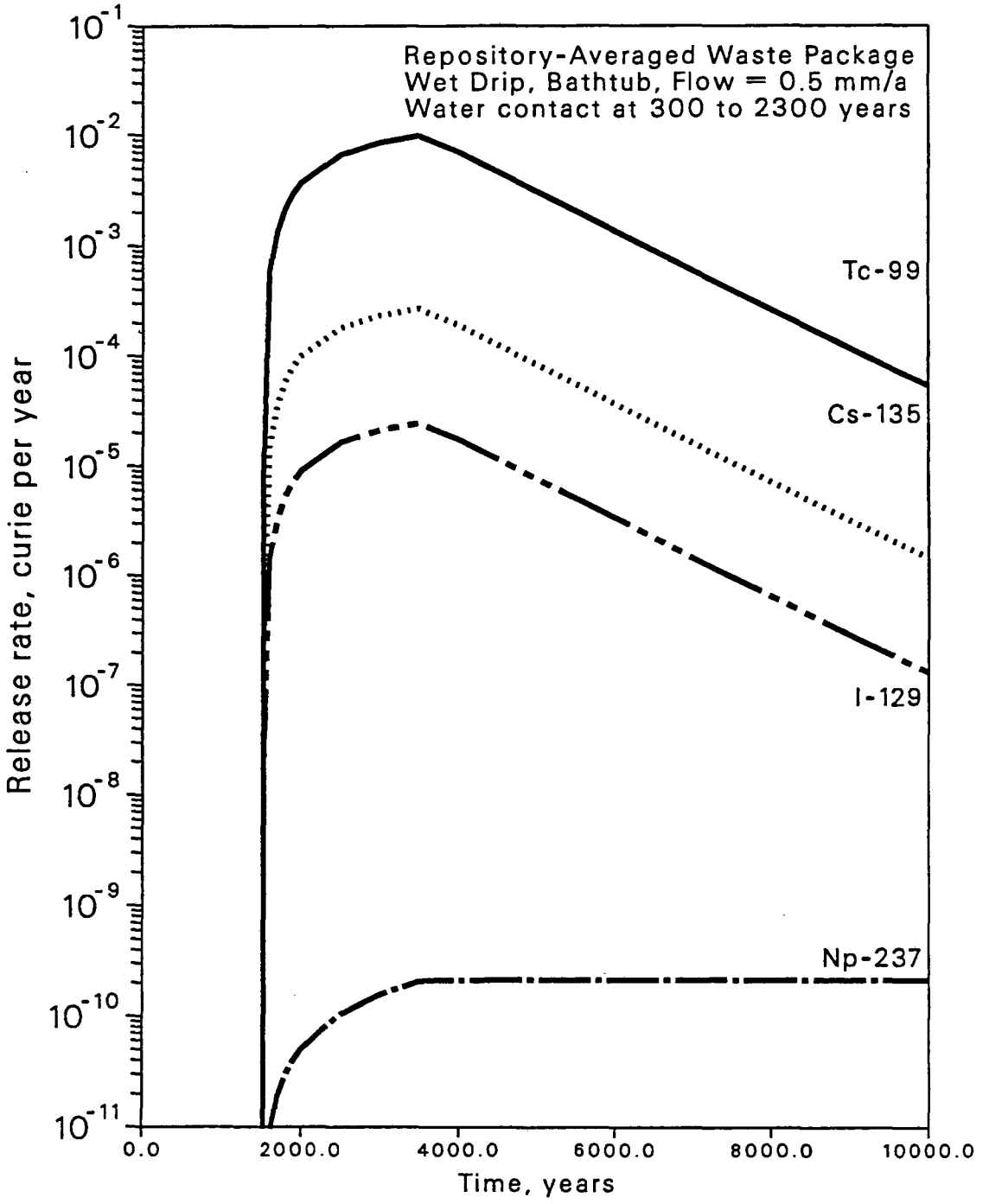

Figure 4.2.3 Release of Tc-99, I-129, Cs-135, & Np-237 in Curies/Year, for a Repository-Average Waste Package Containing Spent Fuel, Wet-Drip Bathtub Contact Mode

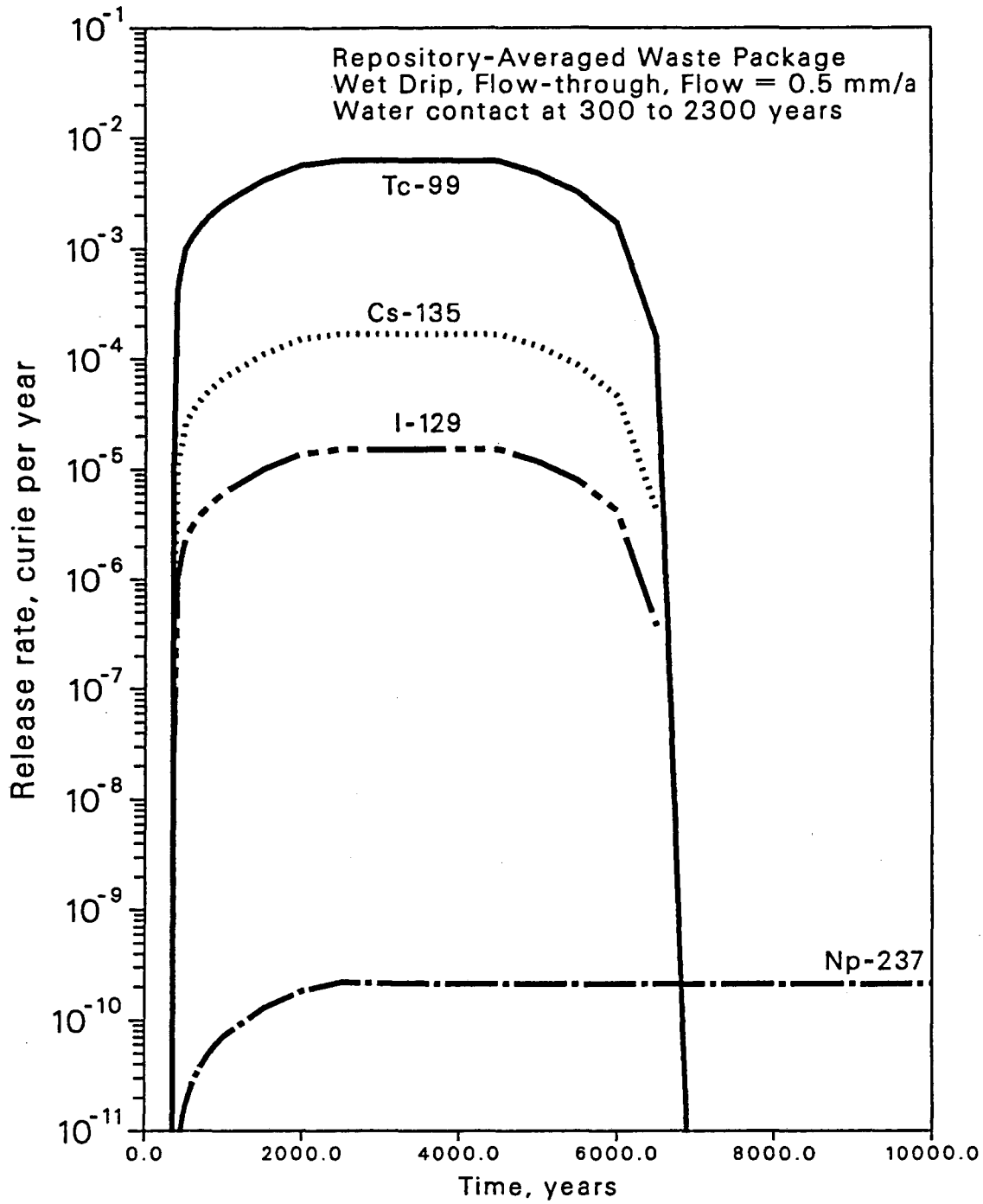

Figure 4.2.4 Release of Tc-99, I-129, Cs-135, & Np-237 in Curies/Year, for a Repository-Average Waste Package Containing Spent Fuel, Wet-Drip Flow-Through Contact Mode

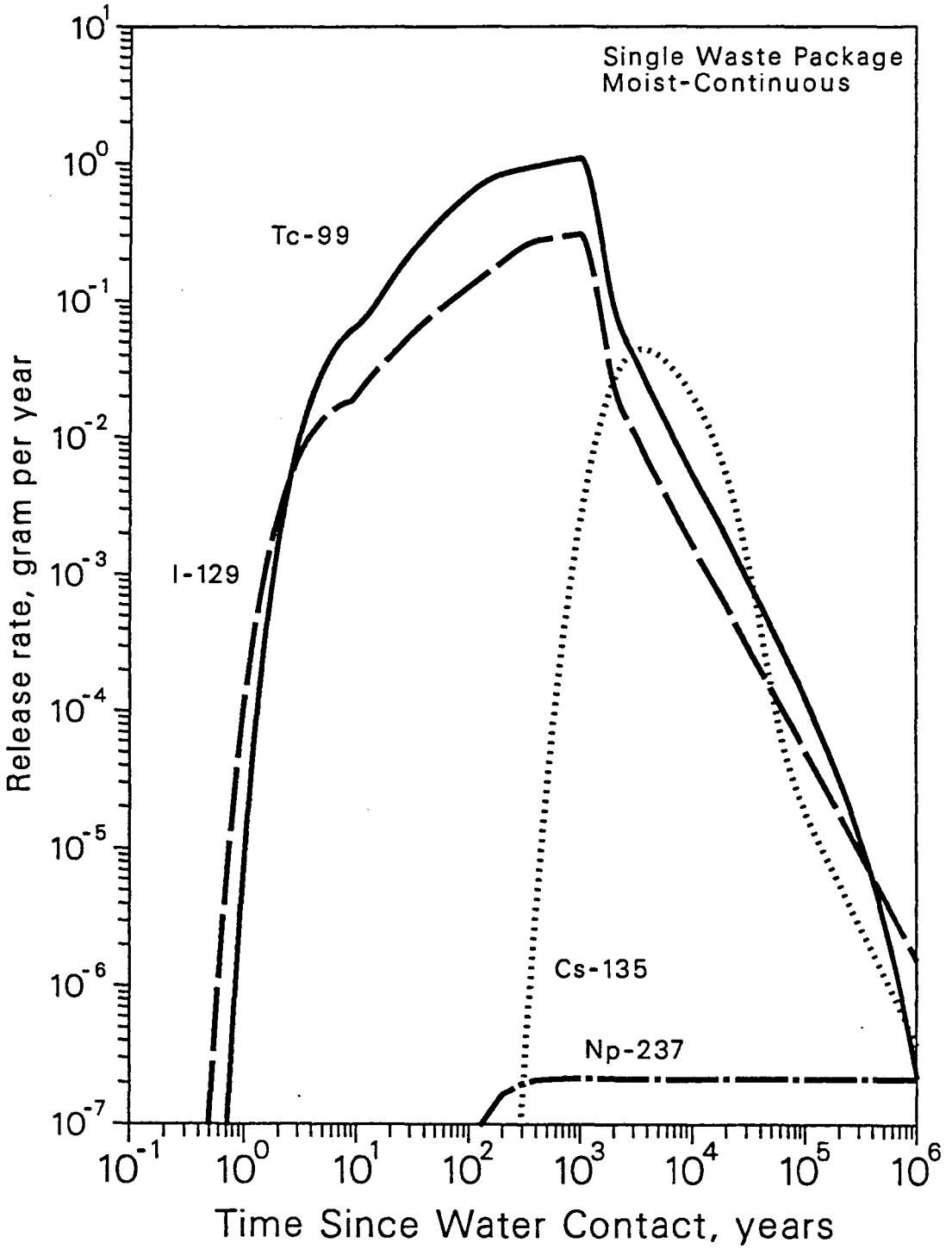

Figure 4.3.1 Matrix Release Rates in Grams/Year for a Single Package Containing Spent Fuel, Moist-continuous Contact Mode

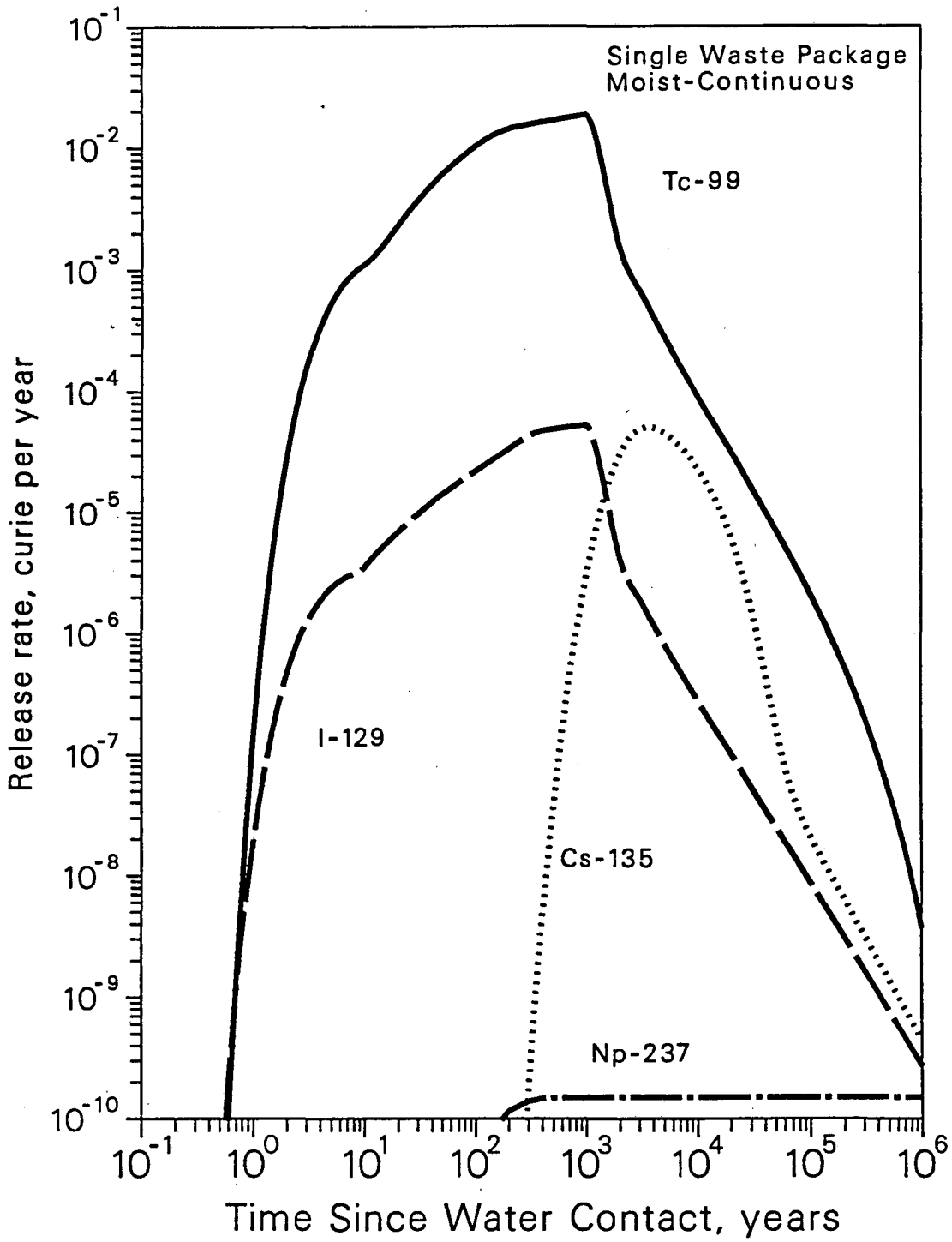

Figure 4.3.2 Matrix Release Rates in Curies/Year for a Single Package Containing Spent Fuel, Moist-continuous Contact Mode

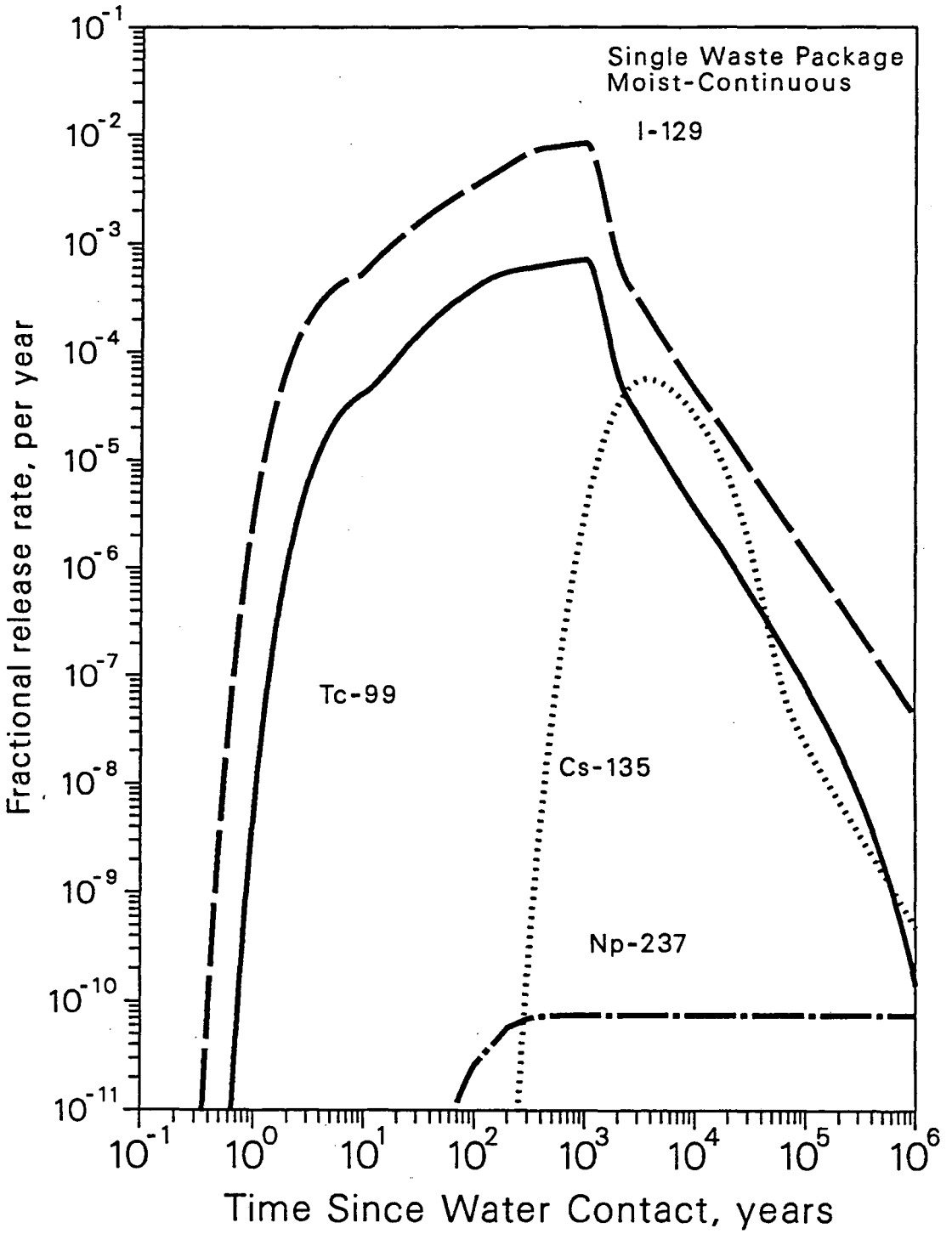

Figure 4.3.3 Matrix Release Rates in Part/Year for a Single Package Containing Spent-Fuel, Moist-continuous Contact Mode

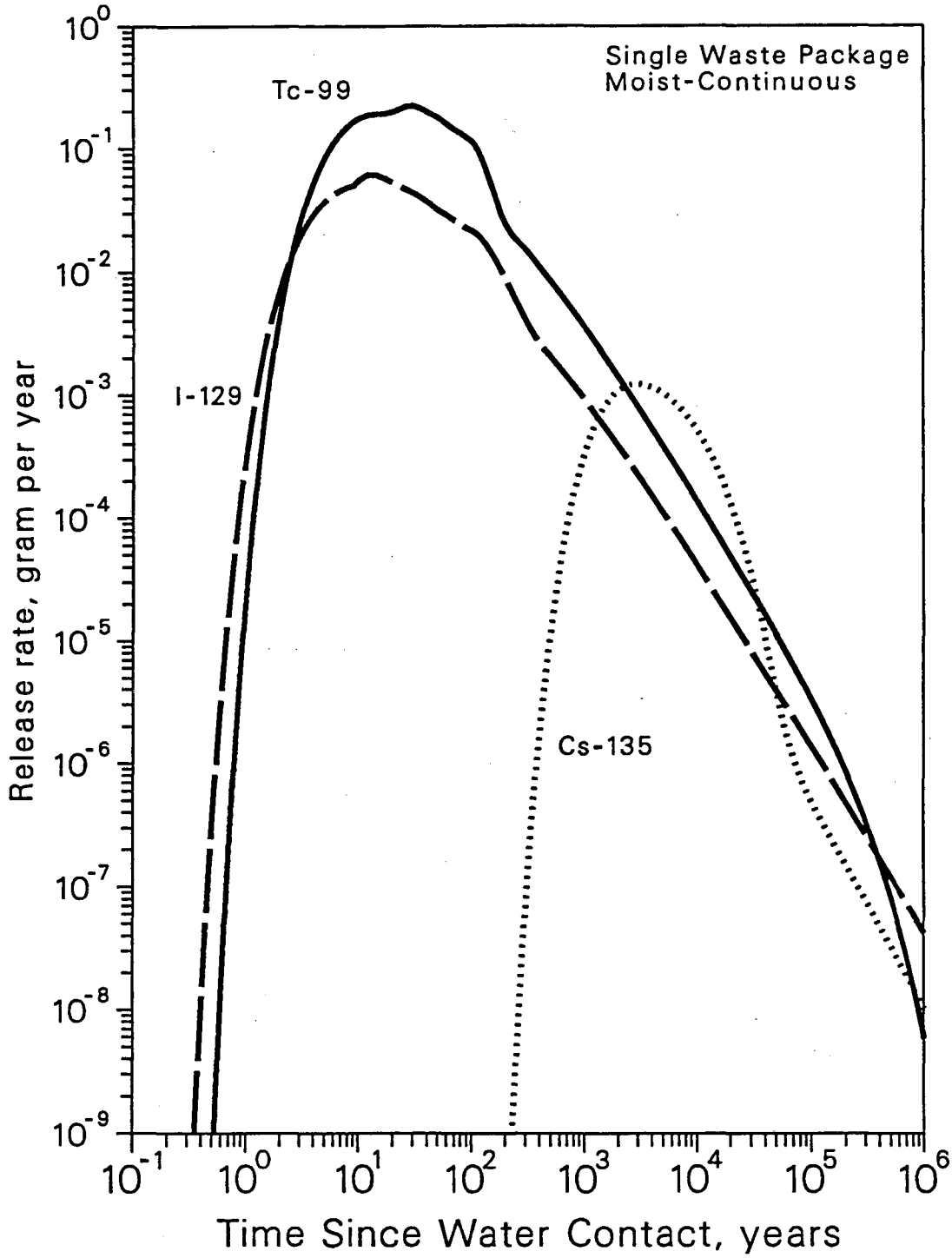

Figure 4.3.4 Gap Release Rates in Grams/Year for a Single Package Containing Spent Fuel, Moist-continuous Contact Mode

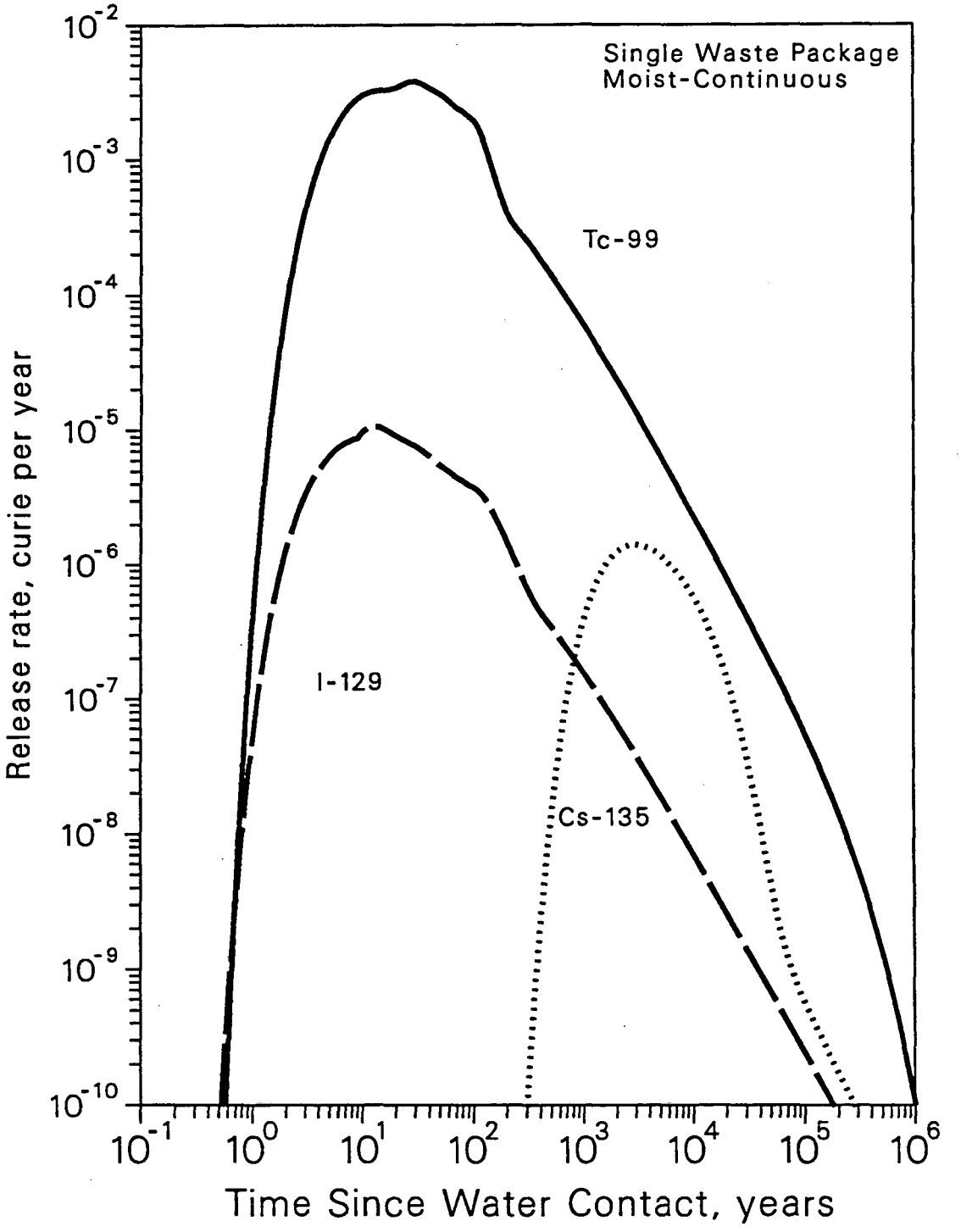

Figure 4.3.5 Gap Release Rates in Curies/Year for a Single Package Containing Spent Fuel, Moist-continuous Contact Mode

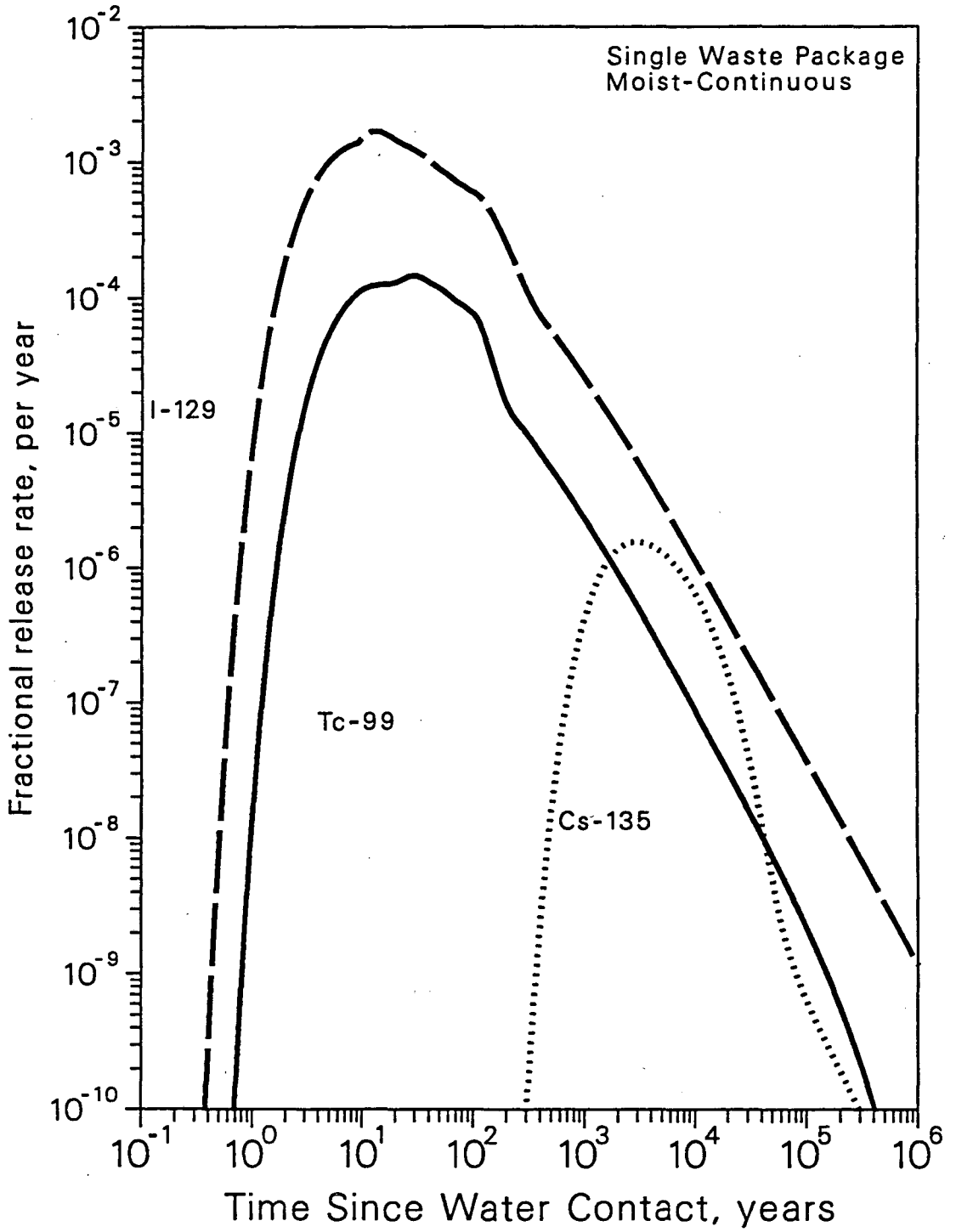

Figure 4.3.6 Gap Release Rates in Part/Year for a Single Package Containing Spent Fuel, Moist-continuous Contact Mode

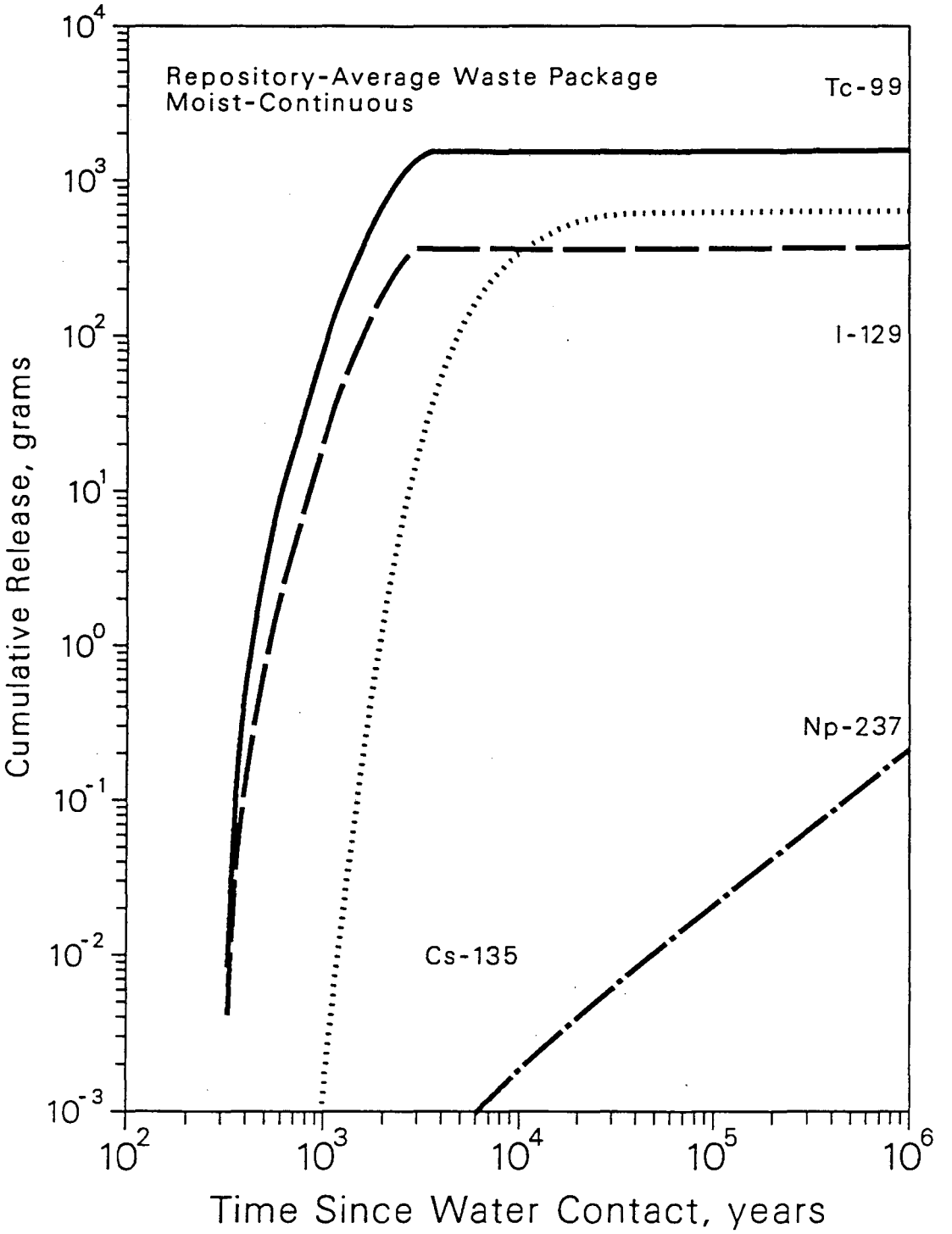

Fig. 4.3.7 Cumulative Release in Grams from the Engineered Barrier System, Moist-continuous

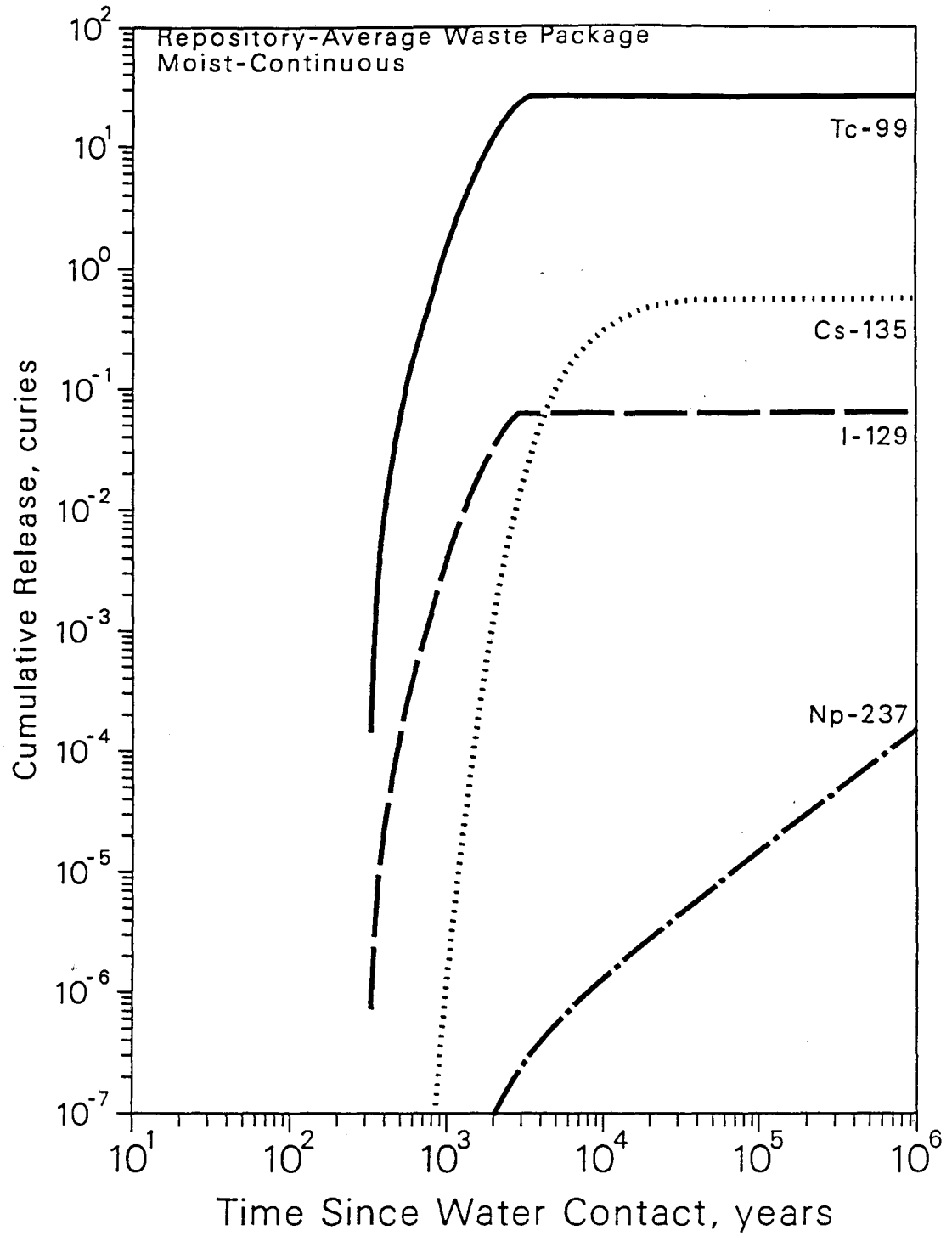

Fig. 4.3.8 Cumulative Release in Curies from the Engineered Barrier System, Moist-continuous

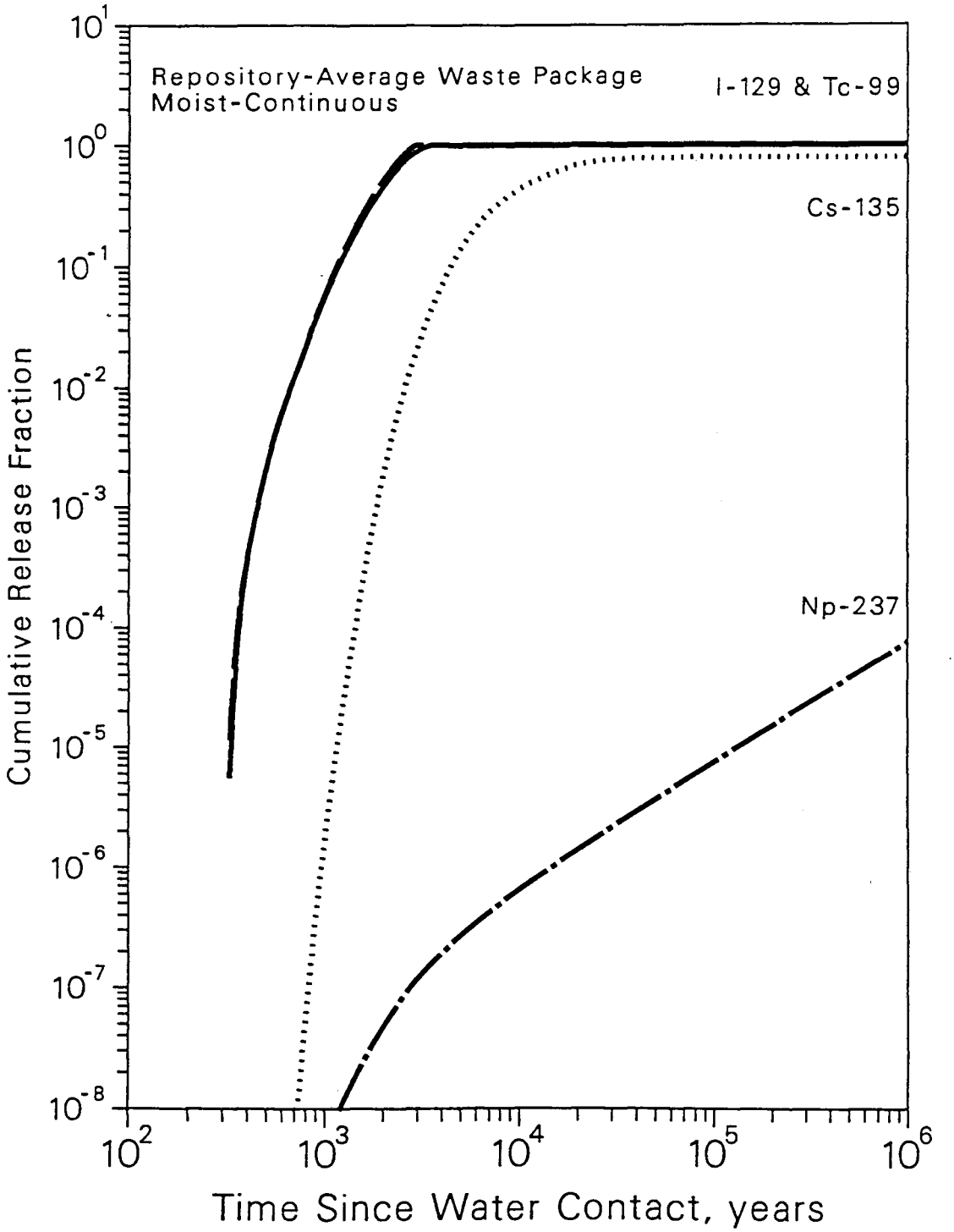

Fig. 4.3.9 Cumulative Release in Fraction (of 1000-Year Inventory) From the Engineered Barrier System, Moist-continuous

App. A. Release Rates, in Ci/a, for a Spent Fuel Package, Wet-Drip Bathtub Contact Mode

	YMP Performance Assessment			
	Single Package - Bathtub			
	Release rate in curies/yr			
Time (yrs)	Tc 99	I 129	Cs 135	Np237
0	0.00E+00	0.00E+00	0.00E+00	0.00E+00
2920	0.00E+00	0.00E+00	0.00E+00	0.00E+00
2921	1.47E-02	3.58E-05	3.92E-04	2.12E-10
3000	1.57E-02	3.83E-05	4.19E-04	2.12E-10
3100	1.69E-02	4.13E-05	4.52E-04	2.12E-10
3500	1.24E-02	3.04E-05	3.33E-04	2.13E-10
4000	8.24E-03	2.02E-05	2.21E-04	2.13E-10
4500	5.46E-03	1.34E-05	1.47E-04	2.14E-10
5000	3.62E-03	8.89E-06	9.73E-05	2.14E-10
5500	2.40E-03	5.90E-06	6.46E-05	2.14E-10
6000	1.59E-03	3.92E-06	4.29E-05	2.14E-10
6500	1.05E-03	2.60E-06	2.85E-05	2.14E-10
7000	6.98E-04	1.73E-06	1.89E-05	2.14E-10
7500	4.62E-04	1.15E-06	1.25E-05	2.14E-10
8000	3.06E-04	7.60E-07	8.32E-06	2.14E-10
8500	2.03E-04	5.05E-07	5.52E-06	2.14E-10
9000	1.35E-04	3.35E-07	3.66E-06	2.14E-10
9500	8.92E-05	2.22E-07	2.43E-06	2.14E-10
10000	5.91E-05	1.48E-07	1.61E-06	2.14E-10
11500	1.72E-05	4.31E-08	4.72E-07	2.14E-10
15000	9.65E-07	2.45E-09	2.68E-08	2.14E-10
20000	1.58E-08	4.06E-11	4.44E-10	2.14E-10
25000	2.58E-10	6.75E-13	7.36E-12	2.14E-10
30000	4.21E-12	1.12E-14	1.22E-13	2.14E-10
35000	6.87E-14	1.86E-16	2.02E-15	2.14E-10
40000	1.12E-15	3.08E-18	3.35E-17	2.14E-10
45000	1.83E-17	5.12E-20	5.56E-19	2.14E-10
50000	2.99E-19	8.49E-22	9.22E-21	2.14E-10
55000	4.89E-21	1.41E-23	1.53E-22	2.14E-10
60000	7.99E-23	2.34E-25	2.53E-24	2.14E-10
65000	1.30E-24	3.88E-27	4.20E-26	2.14E-10
70000	2.13E-26	6.44E-29	6.97E-28	2.14E-10
75000	3.48E-28	1.07E-30	1.16E-29	2.14E-10
80000	5.68E-30	1.77E-32	1.92E-31	2.14E-10
85000	9.28E-32	2.95E-34	3.18E-33	2.14E-10
90000	1.52E-33	4.89E-36	5.27E-35	2.14E-10
95000	2.48E-35	8.11E-38	8.73E-37	2.14E-10
100000	4.04E-37	1.35E-39	1.45E-38	2.14E-10

App. B. Release Rates, in Ci/a, for a Spent Fuel Package, Wet-Drip Flow Through Mode

	YMP Performance Assessment			
	Single Package - Flow Through			
	Release rate in curies/yr			
Time (yrs)	Tc 99	I 129	Cs 135	Np 237
0	0	0	0	0
1750	0	0	0	0
1751	1.66E-02	4.03E-05	4.42E-04	2.19E-10
1760	1.66E-02	4.03E-05	4.42E-04	2.19E-10
1800	6.34E-03	1.51E-05	1.68E-04	2.19E-10
1900	6.34E-03	1.51E-05	1.68E-04	2.20E-10
2000	6.34E-03	1.51E-05	1.68E-04	2.22E-10
2500	6.32E-03	1.51E-05	1.68E-04	2.26E-10
3000	6.30E-03	1.51E-05	1.68E-04	2.28E-10
3500	6.28E-03	1.51E-05	1.68E-04	2.13E-10
4000	6.27E-03	1.51E-05	1.68E-04	2.13E-10
4500	6.26E-03	1.51E-05	1.68E-04	2.14E-10
5000	6.25E-03	1.51E-05	1.68E-04	2.14E-10
5500	6.23E-03	1.51E-05	1.68E-04	2.14E-10
5800	6.23E-03	1.51E-05	1.68E-04	2.14E-10
5801	0.00E+00	0.00E+00	0.00E+00	2.14E-10
6000	0.00E+00	0.00E+00	0.00E+00	2.14E-10
6500	0.00E+00	0.00E+00	0.00E+00	2.14E-10
7000	0.00E+00	0.00E+00	0.00E+00	2.14E-10
7500	0.00E+00	0.00E+00	0.00E+00	2.14E-10
8000	0.00E+00	0.00E+00	0.00E+00	2.14E-10
8500	0.00E+00	0.00E+00	0.00E+00	2.14E-10
9000	0.00E+00	0.00E+00	0.00E+00	2.14E-10
9500	0.00E+00	0.00E+00	0.00E+00	2.14E-10
10000	0.00E+00	0.00E+00	0.00E+00	2.14E-10

App. C. Release Rates, in Ci/a, for a Repository-Average Package, Wet-Drip Bathtub

	YMP Performance Assessment			
	Linear Distribution of Failure - Bathtub			
	Release rate per package in curies/yr			
Time (yrs)	Tc 99	I 129	Cs 135	Np 237
0	0	0	0	0
300	0	0	0	0
1520	0	0	0	0
1521	7.325E-06	1.789E-08	1.959E-07	1.061E-13
1600	5.865E-04	1.432E-06	1.568E-05	8.489E-12
1700	1.360E-03	3.321E-06	3.638E-05	1.910E-11
1800	2.191E-03	5.352E-06	5.862E-05	2.972E-11
1900	2.992E-03	7.307E-06	8.004E-05	4.034E-11
2000	3.736E-03	9.127E-06	9.996E-05	5.097E-11
2500	6.696E-03	1.637E-05	1.793E-04	1.042E-10
3000	8.664E-03	2.119E-05	2.321E-04	1.575E-10
3500	9.969E-03	2.439E-05	2.671E-04	2.109E-10
4000	7.096E-03	1.739E-05	1.904E-04	2.135E-10
4500	4.710E-03	1.156E-05	1.265E-04	2.138E-10
5000	3.121E-03	7.672E-06	8.399E-05	2.140E-10
5500	2.068E-03	5.092E-06	5.574E-05	2.141E-10
6000	1.371E-03	3.379E-06	3.699E-05	2.141E-10
6500	9.084E-04	2.243E-06	2.455E-05	2.141E-10
7000	6.020E-04	1.489E-06	1.629E-05	2.141E-10
7500	3.989E-04	9.882E-07	1.081E-05	2.141E-10
8000	2.644E-04	6.558E-07	7.176E-06	2.141E-10
8500	1.752E-04	4.354E-07	4.763E-06	2.141E-10
9000	1.164E-04	2.898E-07	3.170E-06	2.141E-10
9500	7.863E-05	1.960E-07	2.144E-06	2.141E-10
10000	5.370E-05	1.341E-07	1.466E-06	2.141E-10

App. D. Release Rates, in Ci/a, for a Repository-Average Package, Wet-Drip Flow-Through

YMP Performance Assessment				
Linear Distribution of Failure - Flow Through				
Release rate per package in curies/yr				
Year	Tc 99	I 129	Cs 135	Np 237
0	0.00E+00	0.00E+00	0.00E+00	0.00E+00
300	0.00E+00	0.00E+00	0.00E+00	0.00E+00
350	0.00E+00	0.00E+00	0.00E+00	0.00E+00
351	8.32E-06	2.02E-08	2.21E-07	1.09E-13
400	4.11E-04	9.95E-07	1.09E-05	5.46E-12
500	9.91E-04	2.39E-06	2.63E-05	1.64E-11
600	1.31E-03	3.15E-06	3.47E-05	2.74E-11
700	1.62E-03	3.91E-06	4.31E-05	3.84E-11
800	1.94E-03	4.67E-06	5.15E-05	4.95E-11
900	2.26E-03	5.42E-06	5.99E-05	6.07E-11
1000	2.57E-03	6.18E-06	6.84E-05	7.19E-11
1500	4.15E-03	9.97E-06	1.10E-04	1.28E-10
2000	5.73E-03	1.38E-05	1.53E-04	1.85E-10
2500	6.31E-03	1.51E-05	1.68E-04	2.22E-10
3000	6.29E-03	1.51E-05	1.68E-04	2.20E-10
3500	6.27E-03	1.51E-05	1.68E-04	2.17E-10
4000	6.26E-03	1.51E-05	1.68E-04	2.14E-10
4500	6.25E-03	1.51E-05	1.68E-04	2.14E-10
5000	4.84E-03	1.17E-05	1.30E-04	2.14E-10
5500	3.28E-03	7.95E-06	8.83E-05	2.14E-10
6000	1.71E-03	4.16E-06	4.63E-05	2.14E-10
6500	1.56E-04	3.79E-07	4.21E-06	2.14E-10
7000	0.00E+00	0.00E+00	0.00E+00	2.14E-10
7500	0.00E+00	0.00E+00	0.00E+00	2.14E-10
8000	0.00E+00	0.00E+00	0.00E+00	2.14E-10
8500	0.00E+00	0.00E+00	0.00E+00	2.14E-10
9000	0.00E+00	0.00E+00	0.00E+00	2.14E-10
9500	0.00E+00	0.00E+00	0.00E+00	2.14E-10
10000	0.00E+00	0.00E+00	0.00E+00	2.14E-10

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