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Authors

Pigford, T H
Lee, W W.-L.

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T.H. Pigford and W.W.-L. Lee

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DISCLAIMER

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T. H. Pigford and W. W.-L. Lee

Department of Nuclear Engineering
University of California

and

Earth Sciences Division
Lawrence Berkeley Laboratory
1 Cyclotron Road
Berkeley, California 94720

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WASTE PACKAGE PERFORMANCE IN UNSATURATED ROCK

T. H. Pigford and W. W.-L. Lee
Department of Nuclear Engineering
University of California
and
Earth Sciences Division, Lawrence Berkeley Laboratory
University of California
Berkeley, CA 94720-0001, USA
415 642-6469, 642-5740

ABSTRACT

The unsaturated rock and near-atmospheric pressure of the potential nuclear waste repository at Yucca Mountain present new problems of predicting waste package performance. In this paper we present some illustrations of predictions of waste package performance and discuss important data needs.

1. INTRODUCTION

Here we review the status of methods to predict the performance of high-level waste packages in a repository in unsaturated rock. U.S. regulations require substantially complete containment by the waste container for 300 to 1,000 years. The regulations also limit the release rates of radionuclides from the engineered barrier system for times up to 10,000 years and longer. Reliable quantitative predictions of the waste package performance under these criteria are required. In this paper we discuss techniques for predicting container failure and release rates and give numerical illustrations of results. Uncertainties in these predictions, data needs, and needs for better predictive techniques are also discussed.

2. WASTE PACKAGES AT YUCCA MOUNTAIN

Nuclear waste will be placed in the potential repository at Yucca Mountain in waste packages. Spent fuel assemblies or consolidated fuel rods and borosilicate glass in steel pour canisters will be enclosed sealed containers, probably metallic. Figure 1 is a sketch of typical waste packages, from the *Site Characterization Plan*.[1]

Current design calls for the waste packages to be surrounded by an air gap. Over time the air gap can fill with exfoliated rock and gravel, forming an unintentional backfill. It is also possible to have a layer of engineered backfill around the waste package.

The waste package consists of the waste form, the cladding on spent fuel or the defense-waste pour canister, the outside container, and possibly backfill. These components provide handling and some radiation shielding during the operational phase and containment and corrosion protection in the postclosure period.

3. PERFORMANCE REQUIREMENTS

Although the waste package is generally not seen as the primary barrier for nuclear waste isolation, it must in fact meet specific regulatory requirements. In terms of the **Issues** in the *Site Characterization Plan*, the waste package must meet these postclosure requirements:

Issue 1.4 Will the waste package meet the performance objective for containment as required by 10 CFR 60.113?[2]

Issue 1.5 Will the waste package and repository engineered barrier systems meet the performance objective for limiting radionuclide release rates as by 10 CFR 60.113?[2]

Issue 1.10 Have the characteristics and configurations of the waste packages been adequately established to (a) show compliance with the postclosure design criteria of 10 CFR 60.135,[2] **and (b) provide information for the resolution of the performance issues?**

The waste package must also provide the source term for the evaluation of total system performance or the resolution of

Issue 1.1 Will the mined geologic disposal system meet the system performance objective for limiting radionuclide releases to the accessible environment as required by 10 CFR60.112 and 40 CFR 191.13?[2]

We shall discuss the two main requirements on the waste package.

The Requirement of Substantially Complete Containment

In 10 CFR 60.113(a)(1)(ii)(A), the U. S. Nuclear Regulatory Commission (USNRC) requires that containment

of high-level waste within the waste package be substantially complete for 300 to 1,000 years after closure of the repository. The design and fabrication of large numbers of waste packages to perform for such lengths of time is an unprecedented engineering task. To assure that the performance of the waste packages will actually be as claimed is an even more formidable problem. In the *Site Characterization Plan*, the U. S. Department of Energy is committed to

design the waste packages to provide total containment of the enclosed waste for the containment period under the full range of anticipated repository conditions. [*Site Characterization Plan*, p. 8.3.5.9-5]

The Release-Rate Requirement

In 10 CFR 60.113(a)(1)(ii)(B), the U. S. Nuclear Regulatory Commission requires that the release rate of any radionuclide from the engineered barrier system following the containment period shall not exceed one part in 100,000 per year of the inventory of that radionuclide calculated to be present at 1,000 years following permanent closure. For low-inventory radionuclides, those that constitute less than 0.1 percent of the calculated total curie inventory at 1,000 years, the allowable annual release of a species is a constant value, equal to 10^{-8} of the total curie inventory in the repository at 1,000 years. The calculated release-rate limits for spent fuel are shown in Figure 2, assuming for this illustration that the USNRC requirement applies to individual waste packages. For high-inventory species, the sloping line applies. For low-inventory radionuclides, the horizontal line is the calculated limit. For PWR fuel of 33,000 MW-day/Mg burnup, the total inventory at 1000 years is 1.74×10^3 curies/Mg. For ^{129}I, the 1,000-year inventory is 3.15×10^{-2} Ci/Mg and the fractional release rate limit is obtained by

$(1,740 \text{ Ci/Mg})(10^{-8}/\text{a})/0.0315 \text{ Ci/Mg}$

$= 5.5 \times 10^{-4}$ per year

4. WASTE-PACKAGE ENVIRONMENT

The unsaturated rock and near-atmospheric pressure of the repository horizon at Yucca Mountain present new problems of predicting waste package performance. The major advantage of the Yucca Mountain site is that, with proper placement of waste, the repository will remain dry for many years. To take advantage of this feature, the design is for a high thermal loading in the repository. Figure 3 shows the time-dependent temperature of gas inside a waste package and the waste-container temperature after emplacement in the repository. The entire repository is predicted to remain above the boiling point of water at this altitude for over 1,000 years.

The favorable partially saturated condition is offset by several uncertainties. Three that are mentioned as **Potentially Adverse Conditons** in 10 CFR 60.122 are summaried here.

The partially saturated condition at Yucca Mountain means that air and therefore oxygen can interact directly with waste packages. Thus, in a repository in partially saturated rock, there is an additional release pathway to consider in performance assessment. Moreover, upon loss of containment, oxygen can enter a waste container. Ziracloy cladding on spent fuel may not be an effective barrier against oxygen which can oxidize UO_2, converting it to higher solubility species such as UO_3. If water then enters the waste container, after the thermal period, it can encounter actinides that are readily soluble [10 CFR 60.122(c)(7)].

The heat from the waste will drive the moisture in the partially saturated rock outward in a saturation front. This heat pulse can cause mineral phase changes in the rock [10 CFR 60.122(c)(8)].

"Sealing" of a repository in unsaturated rock is different than for a repository in saturated rock. How does one seal a repository in the vadose zone against gaseous releases? [10 CFR 60.122(c)(20)].

5. PREDICTION OF CONTAINER LIFE

To show compliance with the requirement of substantially complete containment, it will be necessary to predict the timing, size, and frequency of penentrations in waste containers. According to the *Site Characterization Plan*, the reference material for the container at Yucca Mountain is 304L stainless steel [*Site Characterization Plan*, 7-25]. Austenitic materials are generally resistant to uniform corrosion, but localized or stress-assisted forms of corrosion are more likely to be limiting. The projected rates of general corrosion at Yucca Mountain for 304L are [*Site Characterization Plan*, 7-70]

Environment	Corrosion Rate
Unsaturated Steam	0.07 μm/a
Saturated Steam	0.10 μm/a
Water Immersion	0.15 μm/a

No projected rates of localized or stress-assisted corrosion are given in the *Site Characterization Plan*.

Although 304L stainless steel is given as the reference container material in the *Site Characterization Plan*, it is evidently no longer the material of choice. Currently the following are being considered

Copper-Nickel alloy
High-purity copper
Aluminium bronze
825 alloy (high nickel)
316L stainless steel
304L stainless steel

with a choice to be made in April 1990.

With this new round of selection of materials for the waste container, it is timely to seek methods to predict the time and frequency of container failures. Staehle[3] has proposed the use of statistical techniques to project long-term failure rates of containers from short-term experimental data. Figure 4 shows Staehle's illustration of predicted failure rates for Inconel 750, using test data for exposures up to one year, and a Weibell distribution to forecast failure rates to several thousand years.

With such tools in hand to predict the timing and frequency of container failure, we can analyze the flow of gases out of and into a high-level-waste container in the unsaturated tuff of Yucca Mountain.[4] Even though the penetrations due to localized corrosion and cracking may be small, argon gas initially in the hot container can leak out. As the waste package cools, the pressure inside the container can become less than atmospheric, and air can leak in. Here we analyze the flow of gas for various penetration sizes occurring at 300 years.

The *Site Characterization Plan* proposes to define a waste package "failure" as a leak rate of gas of

$$Q = 1 \times 10^{-4} \quad atm - cm^3/s, \tag{1}$$

(ASME Code for Pressure Vessels, Sec. V, Art. 10, App. IV, 1986; [*Site Characterization Plan*, p. 8.3.5.9-35]). This value divided by RT, where R is the gas constant and T the absolute temperature, gives the allowable molar leak rate, above which the container would be considered failed.

Holes and cracks are modeled as a single equivalent hole. The equation describing isothermal, viscous gas flow through a tube of length ℓ and radius r is

$$Q = \frac{\pi r^4 (P^2 - P_0^2)}{16 \mu \ell R T} \tag{2}$$

where Q is the moles of gas flowing per unit time, P and P_0 are the gas pressures at the tube ends, and μ is the gas viscosity. For a waste container containing n moles and volume V of gas, and using the ideal gas law $P = nR\bar{T}/V$

$$\frac{\partial n(t)}{\partial t} = -\frac{\pi r^4 [(nR\bar{T}/V)^2 - P_0^2]}{16 \mu \ell R T_w} \tag{3}$$

where $\bar{T}$ is the average temperature of gas in the container and P_0 is atmospheric pressure.

The initial quantity of gas is given by

$$n(0) = \frac{P(0)V}{RT(0)} \tag{4}$$

Here the average internal temperature $\bar{T}$ and the wall temperature T_w are known functions of time as shown in Figure 3, and μ is a known function of T_w.

Figure 5 shows the calculated gas flow rates as a function of time for various hole radii, assuming that the penetrations occur at 300 years. Also plotted is the allowable flow rate under the leak-rate limit proposed in the *Site Characterization Plan*. Figure 5 shows that the leak-rate limit is not exceeded by a 5-μm hole, and it is exceeded only briefly by larger holes at a fill temperature of 298 K. If the container is filled at the peak gas temperature of 558 K, after the peak gas temperature occurs in the repository, gas can only leak in through a penetration. For holes of 10 μm radius and larger the nearly constant inleakage rate after a few hundred years is determined by the cooling rate and is affected little by hole size. Our analysis of the effect of counter-diffusion appears in a recent publication.

6. PREDICTION OF RELEASE RATES

Table 1 shows the possible release modes for the repository at Yucca Mountain. We will give example calculations of release rates for selected cases, marked with an asterisk. Results of the other cases are in our detailed reports.[5,6]

The unsaturated environment of the tuff repository poses new challenges to predicting waste performance. As long as all waste packages remain surrounded by hot, dry rock, as is predicted for the first thousand or more years, only gaseous radionuclides can be released from failed waste packages. Only after the surrounding rock has cooled sufficiently to allow moisture to exist in the rock matrix can ground water reach the waste packages and penetrate the failed containers. However, prior to moisture penetration air can enter failed containers, as is discussed in Section 5. Air inleakage can oxidize Zircaloy cladding and mobilize more gaseous carbon-14. Oxidation of the uranium dioxide can cause additional cladding failures. The oxidized uranium will be more soluble, and contained radionuclides can be dissolved more readily if later exposed to ground water.

6.1 Release Rate of Gaseous Radionuclides

The gas flow analysis given in Section 5 can give a first estimate of the release rate of gaseous species such as ^{14}C. Assuming that early heating of the waste package

Table 1. Release Modes

Environment	Species			
	Gaseous	Low-Solubility	Instant-Release	Alteration-Controlled
Hot-Dry	√*	No Release	No Release	No Release
Ambient	√	Wet Drip* or Wet Continuous	Wet Drip* or Wet Continuous*	Wet Drip* or Wet Continuous
Saturated	No Release	Wet Continuous	Wet Continuous*	Wet Continuous

volatilizes one percent of the ^{14}C inventory, primarily from the cladding surfaces. penetrations equivalent to a single 5-μm-radius hole and 298 K fill temperature result in an initial argon leak rate of 0.06 mole/a (Figure 5) and a ^{14}C fractional leak rate of 2×10^{-5}/a. For a 10-μm-radius hole. the leak rate would be 16-fold larger. Also from Figure 5 we find that for a 298 K fill temperature and 5-μm holes, about one-fourth of the gas and volatilized radionuclides leak from the container.

6.2 The Wet-Drip Scenario

Even when the surrounding rock has reached an equilibrium moisture content, the repository is not expected to be saturated. The 0.2-m air gap between the container and the surrounding rock is designed to keep the container from contacting the rock. For a scenario to predict the possible magnitude of radionuclide releases to ground water, the Lawrence Livermore National Laboratory assumes that ground water drips from the rock above the waste package and penetrates through cracks and holes in failed containers. If there are no bottom-exit pathways, water can dissolve radionuclides as it slowly fills the container. Assuming that contaminated water can escape from other top penetrations and drip onto the rock below, the release rate after container overflow can be estimated for a low-solubility species by multiplying the volumetric flow rate by the elemental solubility and by the time-dependent atom fraction of the isotope in the element. The Zircaloy cladding is assumed to present no barrier to release of radionuclides to water within the container. Liquid within the container is expected to be well mixed at all times from diffusion and thermal convection. The water flow rate into the failed container is assumed to be the product of the ambient Darcy velocity in the surrounding tuff and the cross-sectional area of the waste package. Smaller or larger flow rates may occur, depending on the extent to which water is channeled transversely in fractures above the waste packages.

The results of Sadeghi *et al.*,[6] calculated for plutonium in spent fuel and borosilicate glass, are shown in Figure 6. The mass release rate of each isotope is normalized to the 1000-year inventory of that isotope. The solubilities are from Bruton's EQ3/6 calculations.[7,8] The estimated void volume within the spent-fuel container is 1.5 m^3. For an estimated Darcy velocity of 0.5 mm/a,[1] the spent-fuel-container fill time is about 8,000 years. Assuming first water penetration at 1,000 years, the first overflow release from the spent fuel container is at 9000 years. The release rate of 6,580-yr Pu-240 decreases rapidly with time because of decay, allowing greater concentrations of the longer-lived Pu-239 and Pu-242 within the constraints of elemental solubility. As 24,400-yr Pu-239 decays, greater solubility-limited concentrations of 379,000-yr Pu-242 are possible, and its release rate increases. The curves are calculated to 100,000 years to illustrate the effect of longer time periods that may be considered. For these parameters, the predicted fractional release rates for plutonium and other low-solubility actinides would fall below the USNRC fractional-release limits, even if those limits were conservatively assumed to apply to individual waste packages rather than to the repository-average release rates.

Results for plutonium released from glass defense waste are also shown in Figure 6. The release begins earlier than for spent fuel because of the smaller void volume of 0.42 m^3 within the defense waste package. The predicted fractional release rate of a species increases with solubility and decreases with increasing inventory. As compared with spent fuel, the lower solubility predicted for plutonium in defense waste overcomes the lower inventory, resulting in a three-fold lower release rate than for spent fuel.

Fractional release rates of readily soluble cesium and iodine that exist in fuel-cladding gap, fuel plenum, and grain boundaries are shown in Figure 7.[6] The liquid in the failed container is assumed to be well mixed at all

times, from diffusion and thermal convection, so the release rate falls off exponentially after the first overflow.

Some of the highly soluble species in the waste matrix may not be release-limited by their solubilities. Laboratory studies suggest that their release may be congruent with the alteration of the waste matrix when it reacts with water, such as conversion of UO_2 in spent fuel to U_3O_7 and the conversion of silica in borosilicate glass to a crystalline mineral phase. Present data are not sufficient to estimate alteration rates, but for illustration we assume a constant mass rate of alteration equivalent to a fractional rate of 0.001/a of the initial inventory of uranium in spent fuel or of silica in defense waste.

The predicted fractional rates[6] of Tc-99 released by waste-matrix alteration are shown in Figure 8 for spent fuel and defense waste. The first liquid overflow is at an average concentration resulting from the slow rise of liquid level in the container. Only at the time of overflow will all of the waste matrix have been exposed to water for alteration and release. The average concentration of long-lived species in the filled container increases further with time, and their release rates increase, until all of the waste matrix has altered and all soluble constituents are dissolved. For the rate constant assumed here, the last waste-solid alteration will be complete 1,000 years after first overflow. The release then decreases exponentially with time as fresh ground water mixes uniformly with the container liquid. These results are only for illustration. Better data are needed to estimate alteration rates appropriate for this calculation.

The release rates of cesium and technetium could be limited instead by solubility. Low-solubility pollucite [$(Cs,Na)_2Al_2Si_4O_{12}\cdot H_2O$] can be formed by cesium and silica from borosilicate glass.[9] Bruton's calculations show that water-waste reactions can result in an increasingly reducing environment[7,8] that can promote a lower solubility of technetium. Also, the iron container is expected not to fail by uniform corrosion but by local penetrations, so iron-water reactions within the container can contribute further to a reducing environment.

The alteration-controlled release rates for borosilicate-glass waste show greater peak values than for spent fuel and decay more rapidly after the peak. This is a consequence of the smaller void volume within the defense-waste container.

6.3 The Wet-Continuous Scenario

The wet-drip scenario assumes that there are no pathways for diffusive release to the surrounding rock. However, part or all of the degraded container can make contact with surrounding rock or with exfoliated rock and rubble that may fill the annular space around the container. Diffusive pathways through the degraded container and into the surrounding rock are possible. We have shown[10] that diffusive mass transfer through the porous media surrounding the waste can result in release rates greater than those estimated for the wet-drip scenario, depending on the magnitude of the diffusion coefficient. Here we show the results of diffusive mass-transfer calculations,[6] and a diffusion coefficient of 3×10^{-2} m^2/a for the annulus and the intact rock.

Figure 9 shows time-dependent fractional release rates of Cs-135 for a ten percent instant release from spent fuel, for various thicknesses of backfill. These results apply for container failure times of up to several thousand years. For a waste package without backfill, the calculated release rate limit is exceeded for less than a decade after container failure. For a 10-cm backfill, the peak release is delayed, of greater duration, and reduced in amplitude. For a 30-cm backfill, the calculated release rate limit is not exceeded.

For these parameters, the release rates are above those predicted for the fuel-alteration wet-drip scenario shown in Figure 7, with no diffusion through the degraded container. However, if the diffusion coefficient is several orders of magnitude below that assumed for the calculation, as may occur if the water in the tuff matrix does not form continuous diffusive pathways, the diffusion-controlled release rates could be less than those from the wet-drip scenario. Also, diffusive resistance from the presence of intact container material has not been considered.

If the continuous diffusive pathways from waste to rock are formed by filling the annular space with exfoliated rock and rubble, the effective diffusion coefficient within this unconsolidated annular layer can be much less than for intact rock. The annulus will not be filled with liquid if the rock never becomes saturated. Diffusive pathways would then occur only where individual rock pieces make physical contact. Sadeghi *et al*[11] have estimated the possible reduction in diffusion coefficient, assuming spherical rubble and using the Hertz equation for contact area between elastic spheres. A large reduction in diffusion coefficient in this region is expected for ambient conditions.

Adopting a 1000-fold reduction in the diffusion coefficient for the 2-cm annular region surrounding the container, we calculate the release rates of Cs-135 into intact rock shown as dashed lines in Figure 9. The release rates are 1000-fold lower than the wet-continuous releases directly into intact unsaturated tuff and are delayed by more than 10,000 years.

The equations for the release rates of soluble species due to waste-matrix alteration are under development.

7. CONCLUSIONS

Waste package performance is an important part of demonstrating compliance with regulatory requirements for the potential geologic repository in unsaturated rock. Some results of analytic prediction of radionuclide release rates have been given. Better data are needed on diffusion coefficients and waste-solid alteration rates.

Our calculation of gas flow through waste-container holes show that leakage through small penetrations need not exceed the proposed failure criteron in the *Site Characterization Plan*, if the internal gas pressure at the time of container closure is properly specified. Prediction of the time, size, and frequency of container failure is needed, to determine substantially complete containment, to calculate release rates, and to derive a source term for total system performance assessment. endorse someone else's untried work in the our paper.

ACKNOWLEDGEMENT

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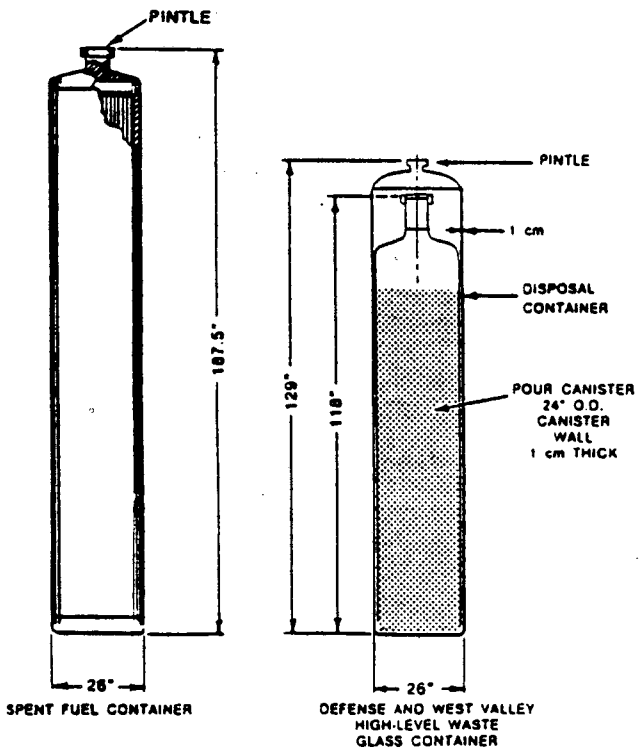

Figure 1. Typical Waste Packages

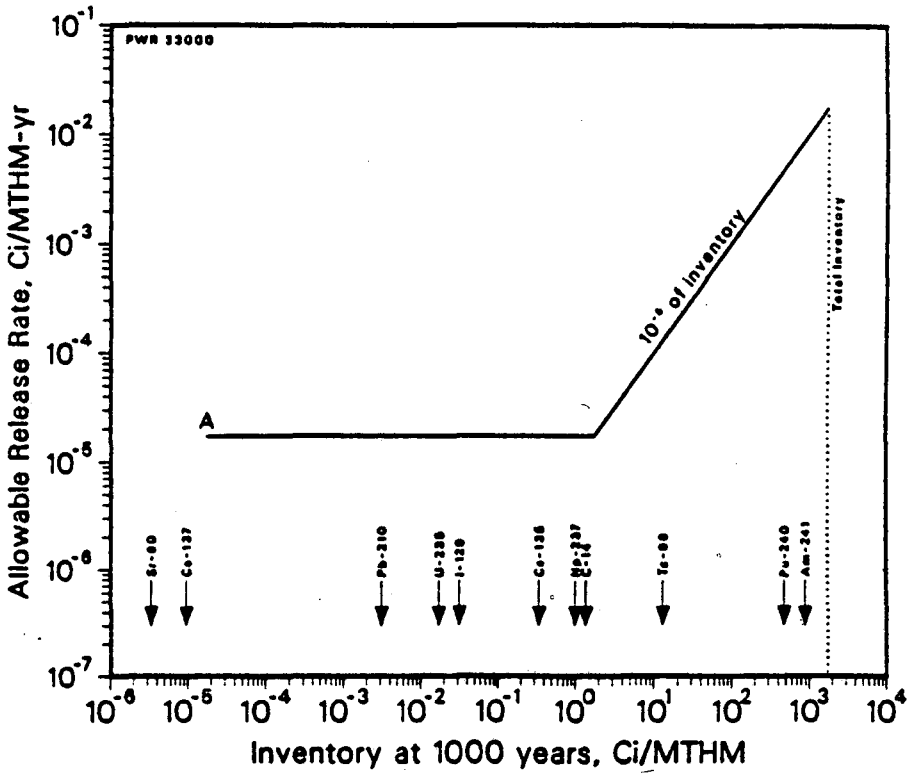

Figure 2. The Release Rate Criterion

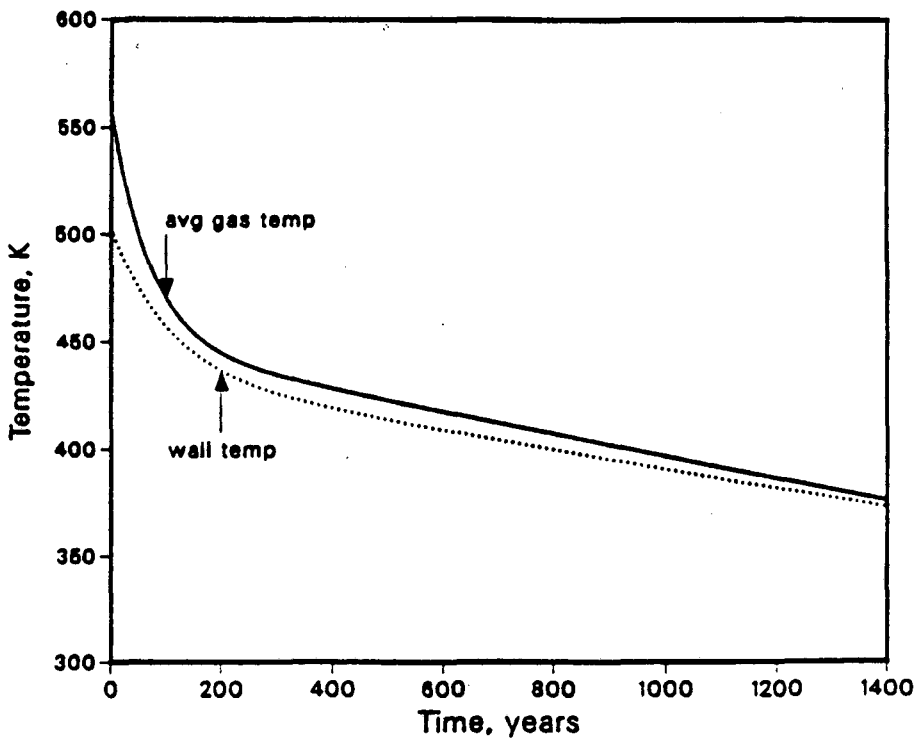

Figure 3. Waste Package Gas and Wall Temperature

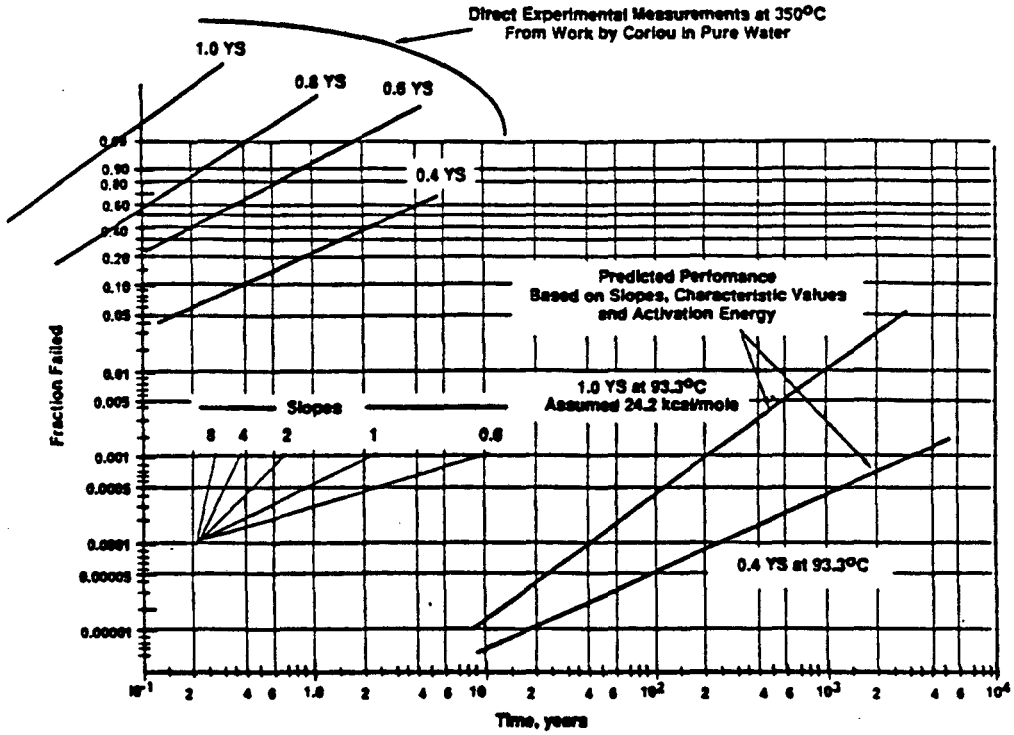

Figure 4. Extrapolation of Container Failure Rates by Weibull Statistics

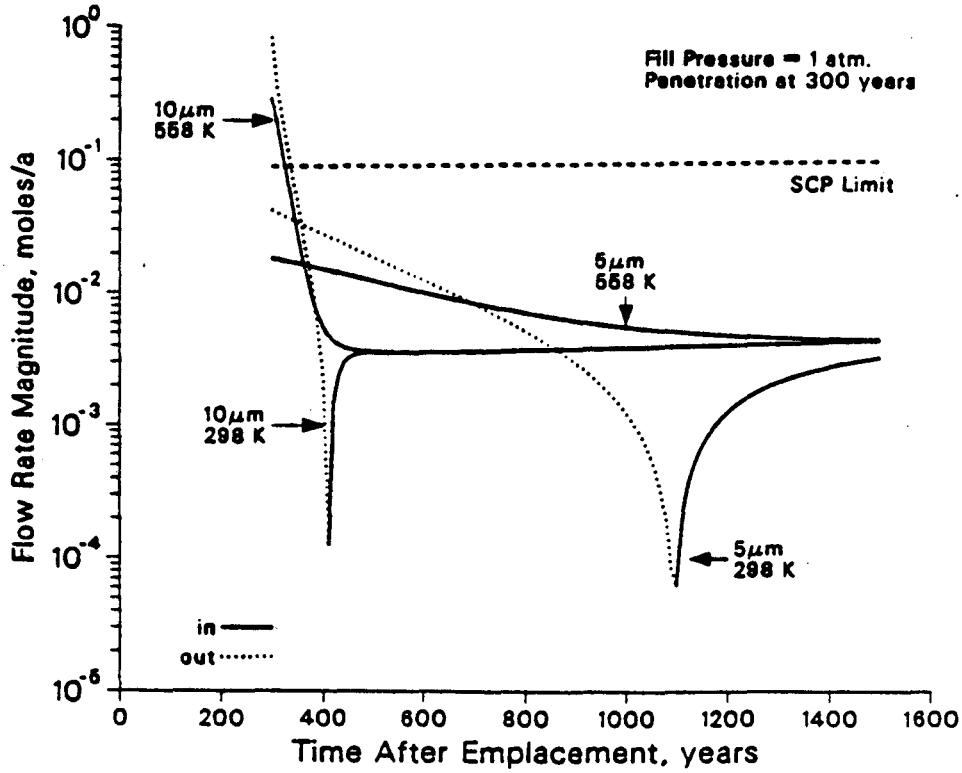

Figure 5. Magnitude of Flow Rate Through a Container Penetration as a Function of Time For Various Apertures and Fill Conditions

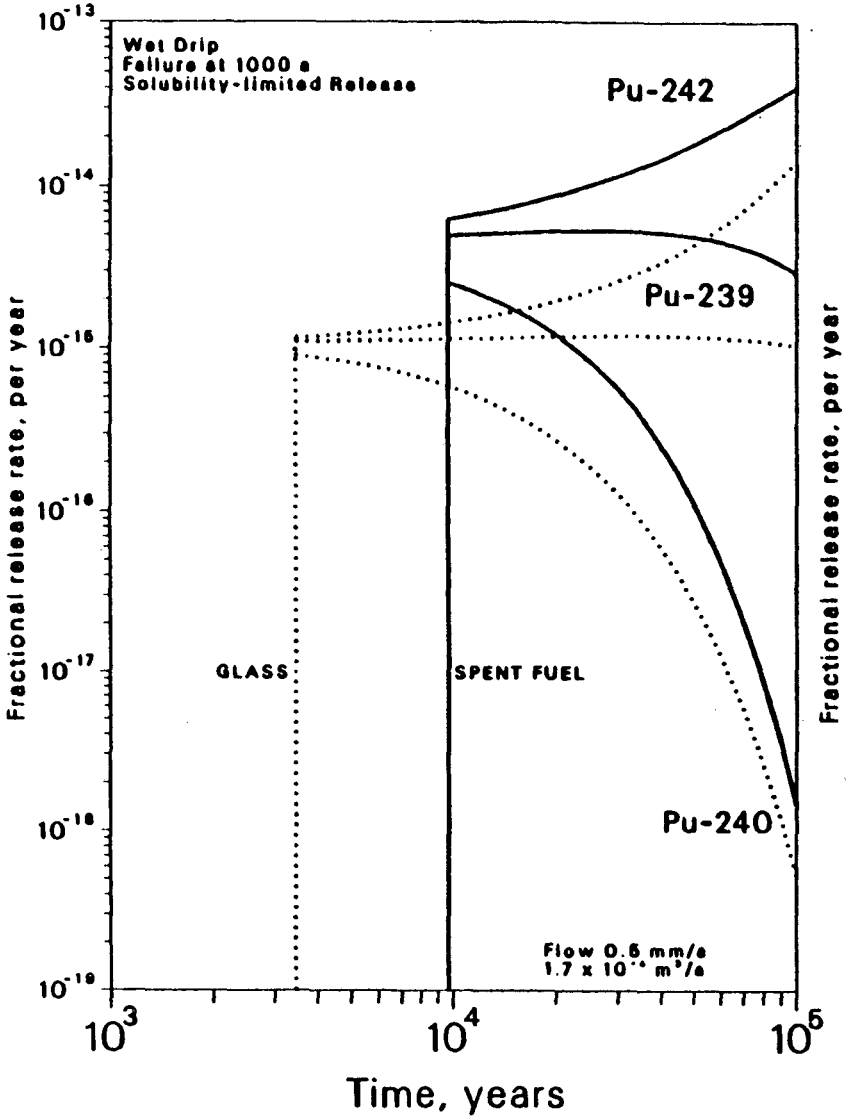

Figure 6. The Wet Drip Fractional Release Rates of Plutonium

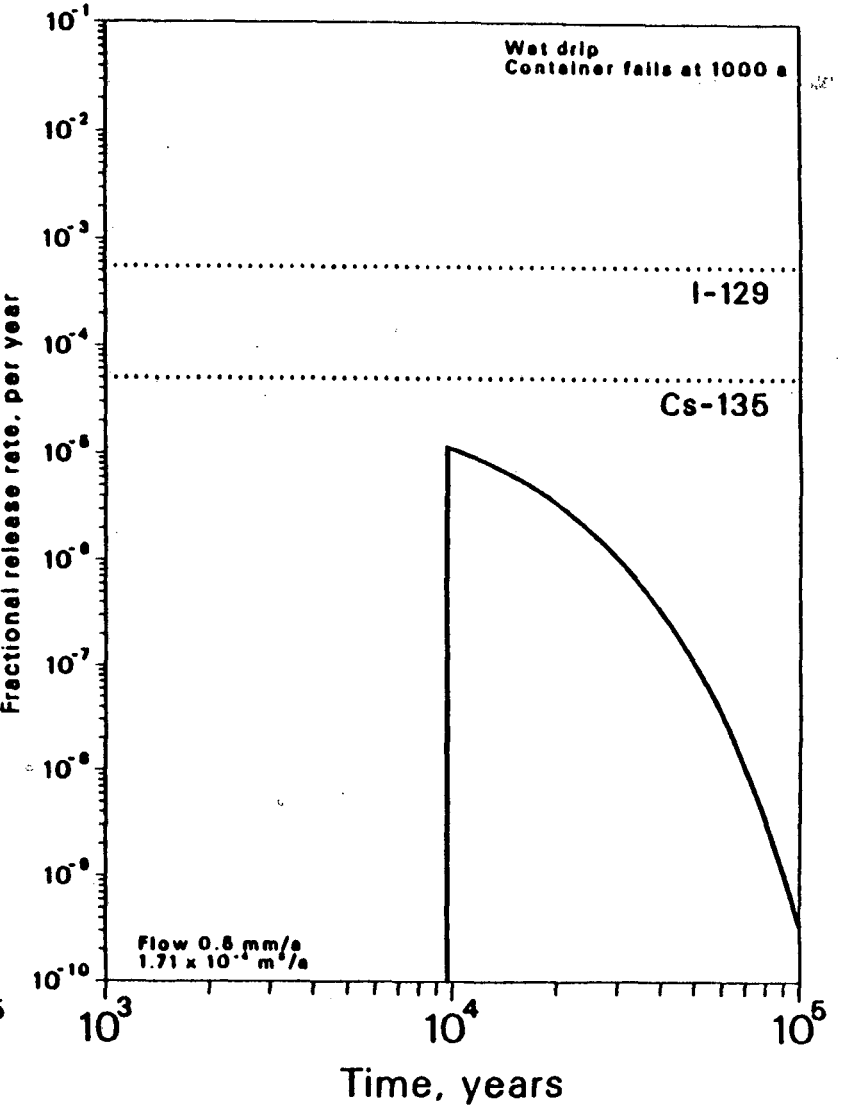

Figure 7. The Fractional Release Rates of Souble Species

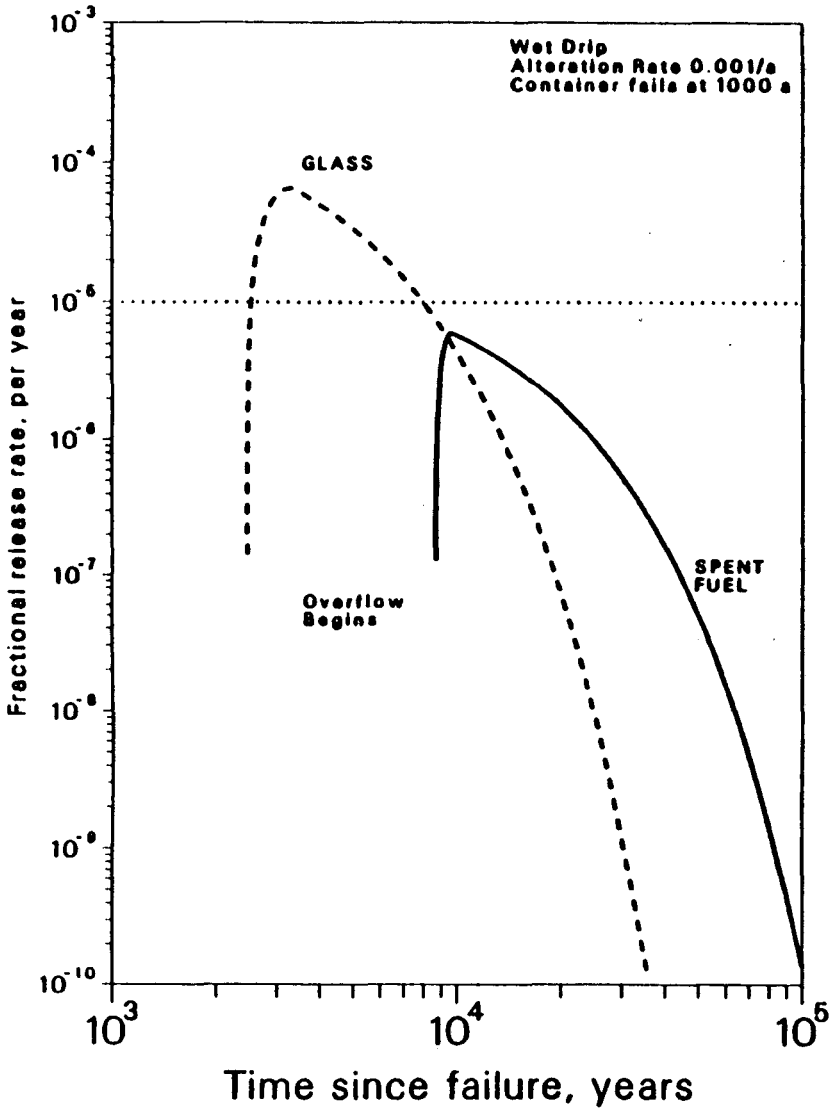

Figure 8. Fractional Release Rates of Tc-99

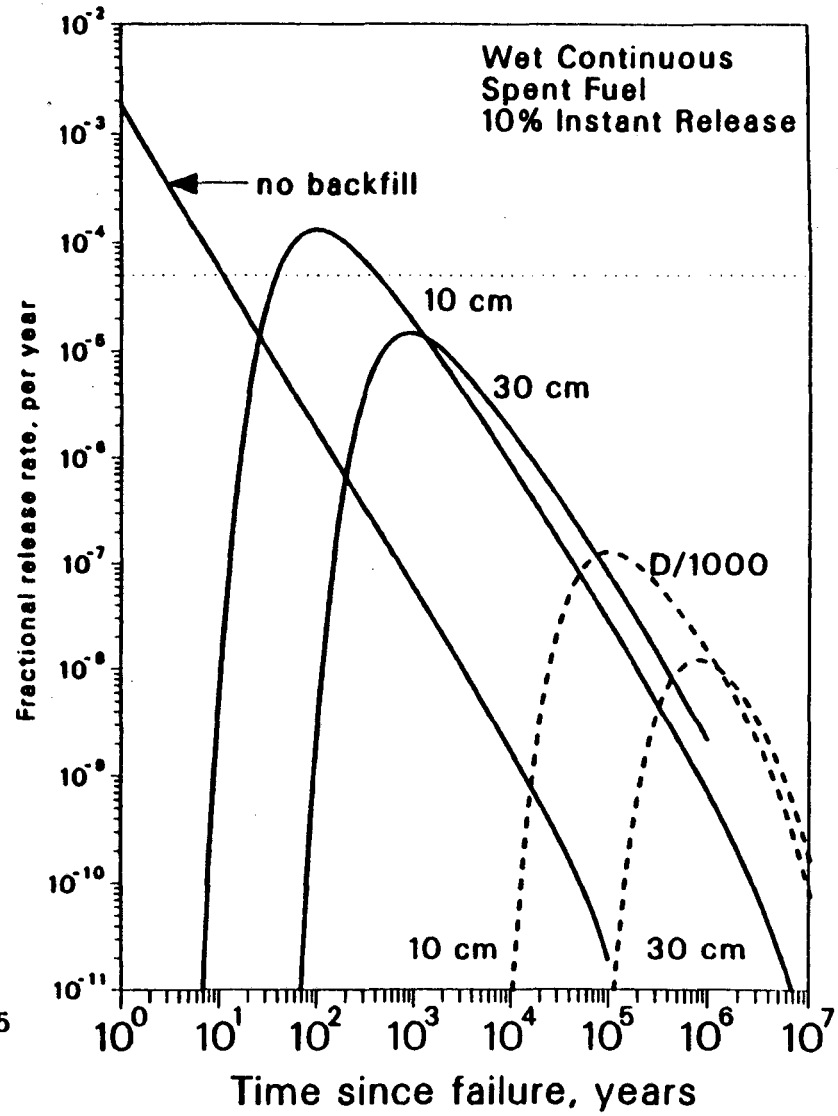

Figure 9. Fractional Release Rate of Cs-135

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