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Authors

Zwahlen, E.D.
Pigford, T.H.
Chambre, P.L.
et al.

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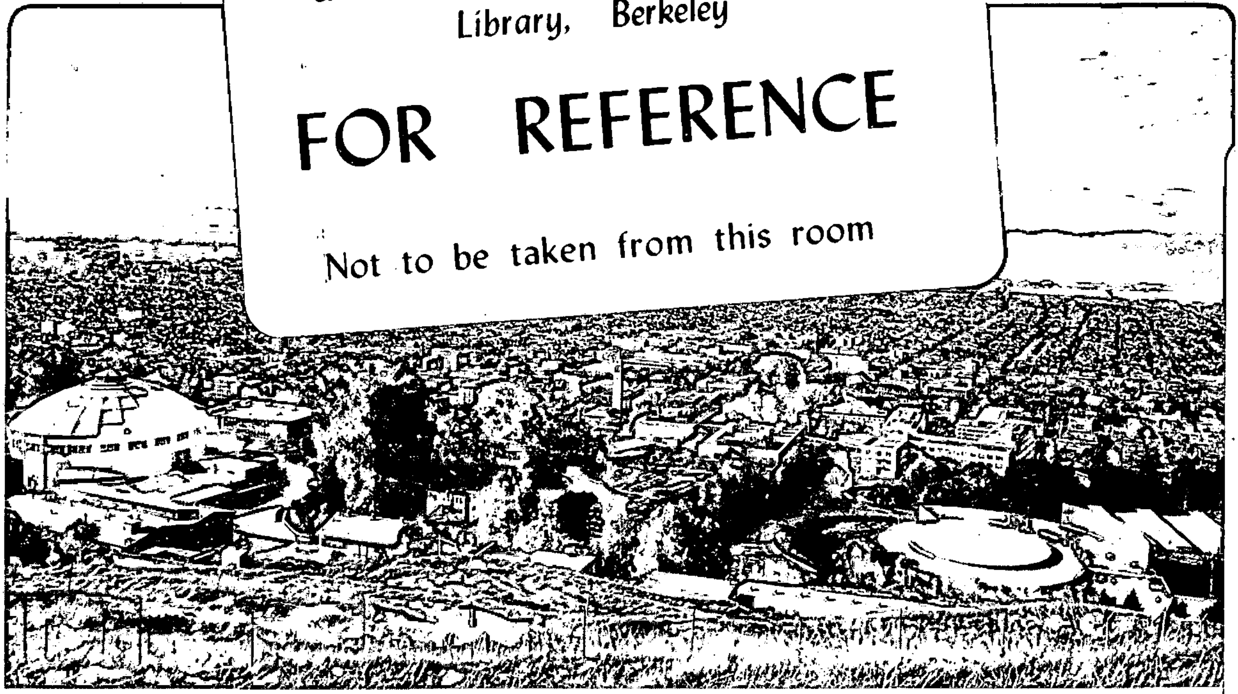

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E. D. Zwahlen, T. H. Pigford, P. L. Chambré, and W. W.-L. Lee

Department of Nuclear Engineering
University of California

Earth Sciences Division
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

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E. D. Zwahlen, T. H. Pigford, P. L. Chambré & W. W.-L. Lee

Department of Nuclear Engineering
University of California
and
Lawrence Berkeley Laboratory, University of California
Berkeley, CA 94720-0001
415 642-6469

1. INTRODUCTION

The potential nuclear waste repository at Yucca Mountain is to be in partially saturated rock. Released radioactive gases would have a direct pathway to the biosphere. The entry of air into spent fuel would promote the oxidation of uranium matrix to more soluble forms. For licensing, the waste package must provide for substantially complete containment of radionuclides, including gases, for 300 to 1,000 years. For safety assessment, a gaseous source term is needed. Thus it is important to study gas flow into and out of nuclear waste containers.

The waste containers will be filled with an inert gas before they are sealed. Due to decay heat, temperature of the container increases. The increased temperature causes the gas pressure inside the container to increase, and inert gas could leak out through a penetration, carrying with it gaseous radioactive material such as $^{14}CO_2$. As the waste cools due to the decay of the heat source, and because of the loss of inert gas through the penetration, the pressure drops. The pressure inside the container will eventually fall below atmospheric pressure. As the waste further cools air can leak in carrying with it oxygen that can volatilize additional ^{14}C and oxidize the fuel.

This paper presents some initial analyses of gas flow into and out of nuclear waste containers, through holes of specified timing and sizes. Also, we predict the release rate of selected volatile species. We investigate the effect of initial temperature and pressure of the container gas and consider the definition of container failure proposed in the *Site Characterization Plan.*[1]

2. BACKGROUND

Some waste packages at Yucca Mountain will contain spent fuel assemblies or consolidated fuel rods; others will contain borosilicate glass in steel pour canisters. Figure 1 is a sketch of typical waste packages, from the *Site Characterization Plan.*

The main requirement on the waste package is

Substantially Complete Containment

The Nuclear Regulatory Commission (USNRC) requires[2] that containment of high-level waste within the waste package be substantially complete for 300 to 1,000 years after closure of the repository. 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 Yucca Mountain Project has proposed in the *Site Characterization Plan* [p. 8.3.5.9-35] to define a waste package as "failed" if the product of pressure and volumetric gaseous leak rate equals or exceeds[3]

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

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.

3. ANALYSIS

We study gas flow through penetrations modeled as a single equivalent hole of radius r and length ℓ, Figure 2. The flow regime can be characterized by the Knudsen number Kn, which is the ratio of the mean free path length λ to the hole diameter, Kn$= \lambda/2r$.

Table I shows the dominant flow regime for various Knudsen numbers.

Table I. Flow Laws for Gases

Kn	Type of Flow
$\gg 1$	Knudsen/Molecular Flow
≈ 1	Slip Flow (Transition)
$\ll 1$	Viscous Flow

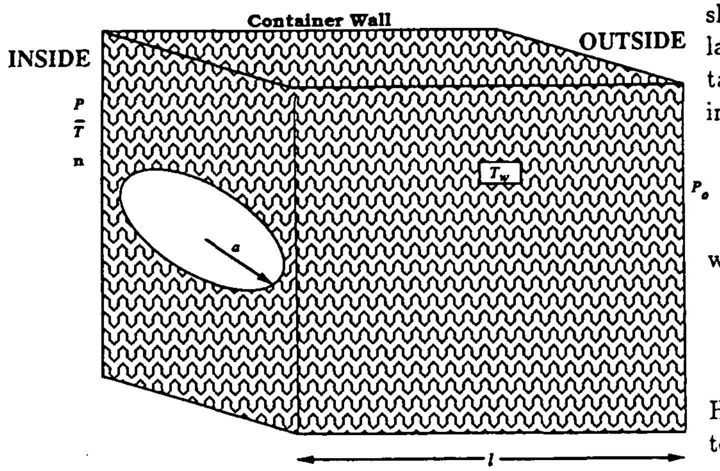

Figure 2. Penetration in a Container Wall

The mean free path is characteristic of the gas and Table II shows some mean free paths of gases of interest.

From Table II, it can be seen that for a single hole of larger than one micrometer in diameter, viscous flow will dominate. We will show later that gas flow through holes of less than one micrometer in diameter will be negligible, under any flow law.

Table II. Mean Free Paths of Selected Gases

Gas	λ at 298 K	λ at 500 K
Air	6.6×10^{-8} m	1.1×10^{-7} m
Argon	6.9×10^{-8} m	1.2×10^{-7} m
CO_2	4.3×10^{-8} m	7.3×10^{-8} m
Krypton	5.3×10^{-8} m	8.9×10^{-8} m
Xenon	3.9×10^{-8} m	6.5×10^{-8} m

Source: Calculated using data from Reference 4.

Viscous Flow Model

The equation for steady viscous or Poiseuille flow of an ideal gas in a tube at constant temperature is

$$Q = \frac{\pi r^4 (P^2 - P_0^2)}{16 \mu \ell R T} \tag{1}$$

Here Q is the mass of gas flowing per unit time, P and P_0 are the gas pressures inside and outside the container, and μ is the absolute viscosity of the gas. Here we assume that the penetrations through the container wall of thickness ℓ can be approximated by a single equivalent hole of radius r and length ℓ, with the gas in local thermal equilibrium with the container wall at temperature T_w. Because the temperature in the repository changes slowly, over hundreds of years, the steady-state gas flow law is applicable. In a waste container of volume V containing n moles of gas, the equation can be transformed into a differential equation in terms of n

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

with the initial quantity of gas given by the ideal gas law

$$n(0) = \frac{P(0) V}{R T(0)} \tag{3}$$

Here the average internal temperature $\bar{T}$, and the wall temperature T_w, are known functions of time, and μ is an empirical function of T_w. $\bar{T}$ is approximated as the arithmetic average of the peak fuel cladding temperature and T_w. The time for the waste to heat to its maximum temperature after emplacement is neglected.

Knudsen/Molecular Flow

For extremely small holes, molecular flow applies

$$Q = \frac{8 \pi r^3 (p - p_0)}{3 \ell \sqrt{2 \pi m R T}} \tag{4}$$

where m is the gas molecular weight, and p and p_0 are the partial pressures of each gaseous species.

Counter-Diffusion Model

Fill gas may leak out through a penetration, followed by inleakage of air as the waste package cools. Even though air is flowing inward, gaseous radionuclides can escape by diffusion against the flow. In particular we analyze the diffusion of $^{14}CO_2$ against the inward movement of air.

We analyze counter-diffusion release through the equivalent tubular penetration, with quasi-steady-state flow and diffusion. The geometry is the same as Figure 2. Assuming slug flow and constant-concentration boundary conditions at the tube ends, the steady-state mass balance equation for the radionuclide concentration c is

$$\bar{v} \frac{dc}{dz} = D \frac{d^2 c}{dz^2}, \qquad 0 \le z \le \ell \tag{5}$$

with boundary conditions

$$c(0) = c_0 = M/V \tag{6}$$

$$c(\ell) = 0 \tag{7}$$

Here z is the direction of flow along the tube, M is the initial inventory of volatilized radionuclide in the container, D is the gas diffusion coefficient, and $\bar{v}$ is the gas velocity in the tube averaged over the tube cross-section. For counter-current diffusion, $\bar{v}$ is negative. The use of the boundary conditon (7) ensures the maximum amount for diffusion, a conservative estimate.

By solving (5) for c we obtain the mass transfer rate $\dot{m}$ as

$$\dot{m} = \pi r^2 \left(\bar{v}c - D\frac{dc}{dz} \right) \tag{8}$$

and we define the fractional release rate f as

$$f = \dot{m}/c \tag{9}$$

The fractional release rate is

$$f = \begin{cases} \pi r^2 \bar{v} \dfrac{e^\alpha}{e^\alpha - 1}, & \bar{v} <> 0; \\ \pi r^2 D/\ell, & \bar{v} = 0 \end{cases} \tag{10}$$

where $\alpha = \bar{v}\ell/D$.

4. NUMERICAL ILLUSTRATIONS

Viscous Flow

We integrate (3) numerically to calculate the quantity n of gas inside a container and the gas flow rate in or out as functions of time. $\bar{T}$ and T_w have been obtain from *Site Characterization Plan*, [p. 7-41] and extrapolated (Figure 3). Argon viscosity is used when gas flows out, and air viscosity is used when air flows in. The results will be presented for two hole sizes, 5×10^{-6} meters (5μm) and 10μm, and two fill temperatures, 298 K and 558 K. The fill pressure is 0.1 MPa, and the hole is assumed to occur at emplacement or 1000 years after the waste is buried. Figures 4 and 6 show the mass of gas inside the waste container as a function of time, and Figures 5 and 7 show the gas flow rate through the hole as a function of time.

In Figure 4 the dotted lines refer to outleakage and the solid lines refer to inleakage. For penetration at emplacement, a 5-μm hole, and a fill temperature of 298 K, argon slowly leaks out until about 800 years, after which the leaking and repository cooling cause the internal pressure to fall below atmospheric, and air leaks in. For a 5μm hole, 25% of the argon flows out, while for a 10μm hole, 32% flows out. For a 10-μm hole, the internal pressure rapidly falls to atmospheric, and inleakage begins at about 70 years. For a 558 K fill temperature, the maximum gas temperature in waste packages, there is no thermally generated buildup of pressure and thus no gradient for outleakage. A penetration results only in inleakage of air because the internal pressure is always less than atmospheric. Although not shown, curves for holes larger than 10-μm are nearly identical to the 10-μm curves. Thus, for these large holes, the quantity of gas depends only on the rate of cooling, independent of hole size.

Figure 5 shows the flow rate of gas in and out of the waste container for penetration at emplacement, the derivative of the curves in Figure 4. The dashed curve labeled "SCP limit" is the maximum allowable flow rate, above which the container is considered failed. The slight upward slope is due to the decreasing wall temperature. For a 298 K fill temperature, the flow rate through a 5-μm hole never exceeds this limit, but the flow rate through a 10-μm hole briefly exceeds it. For a 558 K fill temperature, the pressure gradient and flow are zero at the time of penetration. As the waste cools, the rate of inleakage increases, reaches a maximum, and then decreases to a nearly constant rate that depends only on the cooling curve. Again, the slight upward slope at late times is because T_w in the denominator of (2) is decreasing with time.

The results above are important in assessing waste container performance, but we are also interested in radionuclide release rates.[5] Figures 4 through 8 can be used to estimate release rates. In Figure 4 for a 298 K fill temperature and a 5-μm hole, about one-fourth of the gas and volatilized radionuclides leaks from the container. For 10-μm holes or larger, almost half of the gas leaks out before air inleakage begins.

If we assume that $^{14}CO_2$ is leaked out at the same rate as argon, and we know its volatile fraction, then we can compute a $^{14}CO_2$ molar flow rate and fractional leak rate. The molar flow rate of $^{14}CO_2$ is the molar flow rate of argon from Figure 5, times the mole fraction of $^{14}CO_2$. A fractional leak rate of argon can be computed as the leak rate from Figure 5 divided by the quantity of gas in the container from Figure 4.

The fractional leak rate for other volatile species is the ratio of the argon leak rate to the argon mass in the container times the volatile fraction of species.

Assuming that heating of the waste package volatilizes 1 percent of the ^{14}C inventory, primarily from cladding surfaces, a 5-μm hole and 298 K fill temperature result in an initial argon leak rate of 0.06 mole/yr and a ^{14}C fractional leak rate of 2×10^{-5}/yr. For the 558 K fill temperature, this model predicts that no radioactive material will be released. Here counter-diffusion is neglected.

We have also studied the effect of initial fill pressureand fill volume, which along with the fill temperature gives the initial inert gas inventory. Qualitatively the curves are the same as Figures 3-7 for 1 atm. For fill pressures greater than 1 atm., the fractional release rates are greater than for 1 atm. For 2 atm. and a 5-μm hole, about 60 percent of the volatile radionuclides is released with the outflow. For 10-μm holes or larger, 70 percent is released. For higher initial pressures, the time of air inleakage time is delayed a few hundred years for 5-μm holes, but it is not affected for 10-μm holes or larger. If the pressure at penetration is less than 1 atm., air leaks in immediately. Pressurizing the container increases the release of volatile radionuclides, but an initial vacuum allows more air to leak in.

Knudsen/Molecular Flow

To show that the gas flow rate will be negligible in the range where molecular flow is dominant, we calculate the flow rate of argon at 500 K through various aperture sizes, when identical pressure gradient applies ($P = 0.19$ MPa, $P_0 = 0.1$ MPa). The results are shown in Table III.

Table III. Comparison of Molecular and Viscous Flows for the Same Pressure Gradient

Flow Rate (moles/a)		
Hole Diameter μm	Molecular	Viscous
0.01	1.6×10^{-10}	1.1×10^{-12}
0.1	1.6×10^{-7}	1.1×10^{-8}
1	10^{-4}	10^{-4}
10	10^{-1}	10
100	10^{2}	10^{4}

Table III shows that while the molecular flow law will predict a flow rate higher than the viscous flow law for extremely small holes, the actual quantity of gas moving through the very small penetrations is miniscule.

We also calculate the importance of counter-diffusion. Because counter-flow is a resistance to diffusion, assuming $\bar{v} = 0$ will give an upper bound to the counter-diffusion release rate. The upper curve in Figure 8 shows the diffusive fractional release rate as a function of penetration radius for D=0.27 cm^2/s, the diffusion coefficient for CO$_2$-air at 400 K. Multiplying these values by the fraction of the total radionuclide inventory that is volatilized, gives the inventory-based fractional release. For a 5-μm hole and 1 percent ^{14}C volatilized, the diffusive fractional release rate is 7×10^{-8} year^{-1} based on total ^{14}C inventory.

The lower curve in Figure 8 shows the fractional release with a molar flow rate of 5×10^{-3} moles/year, which is the late-time inflow rate from Figure 5. This corresponds to a volumetric flow rate, $\pi r^2 \bar{v}$, of 5×10^{-6} cm^3/s at 390 K. For holes smaller than 30-μm there is a reduction in the release rate, but for larger holes the flow is slow and diffusion is the dominant release mode.

5. CONCLUSION

We have studied gas flow out of and into a waste container for various fill conditions and holes sizes. We developed some first-cut methods of calculating gas flows for holes and made some estimates of source terms of gaseous radionuclides.

For holes of 5-μm or less, the flow rates are much less than the SCP-failure criterion. For holes 10-μm or larger, the long-term flow rates are controlled by the cooling rate and are about the same as for 10-μm holes. We found the flow rate through a 5-μm hole is below the SCP leak rate criterion, but for 10-μm holes or larger the leak rate is exceeded briefly. For holes 30-μm or larger, counter-current diffusive release is important because the resistance due to inflow of air is small.

We also obtained some insights for designers, such as whether to pre-pressurize and preheat the waste containers at assembly.

ACKNOWLEDGEMENT

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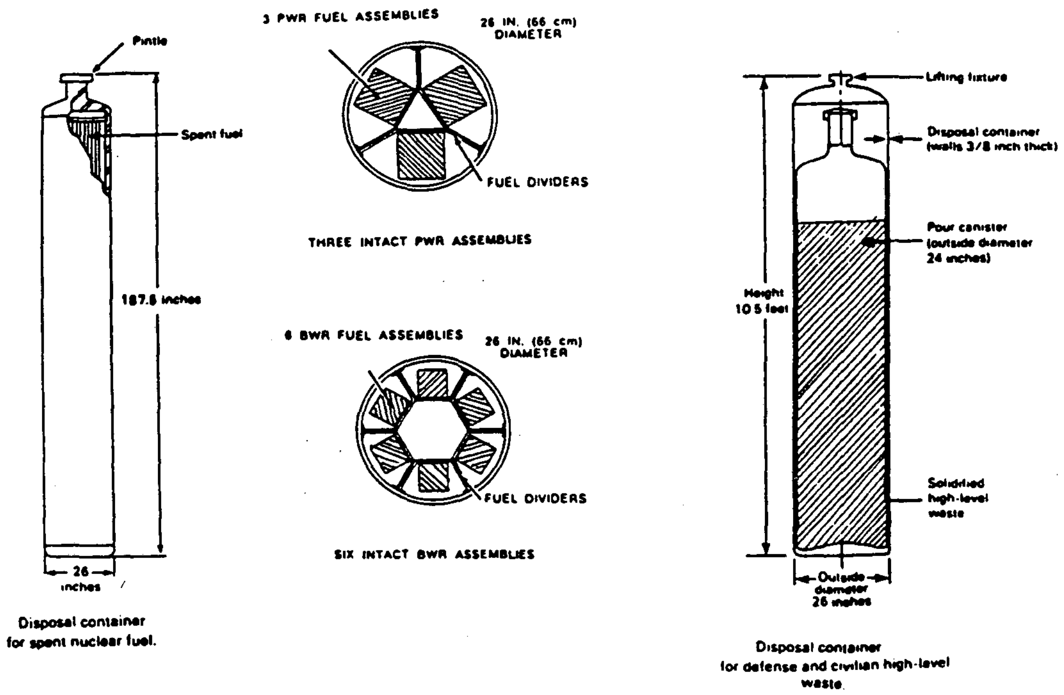

Disposal container
for spent nuclear fuel.

THREE INTACT PWR ASSEMBLIES

SIX INTACT BWR ASSEMBLIES

Disposal container
for defense and civilian high-level
waste.

Figure 1. Typical Waste Packages

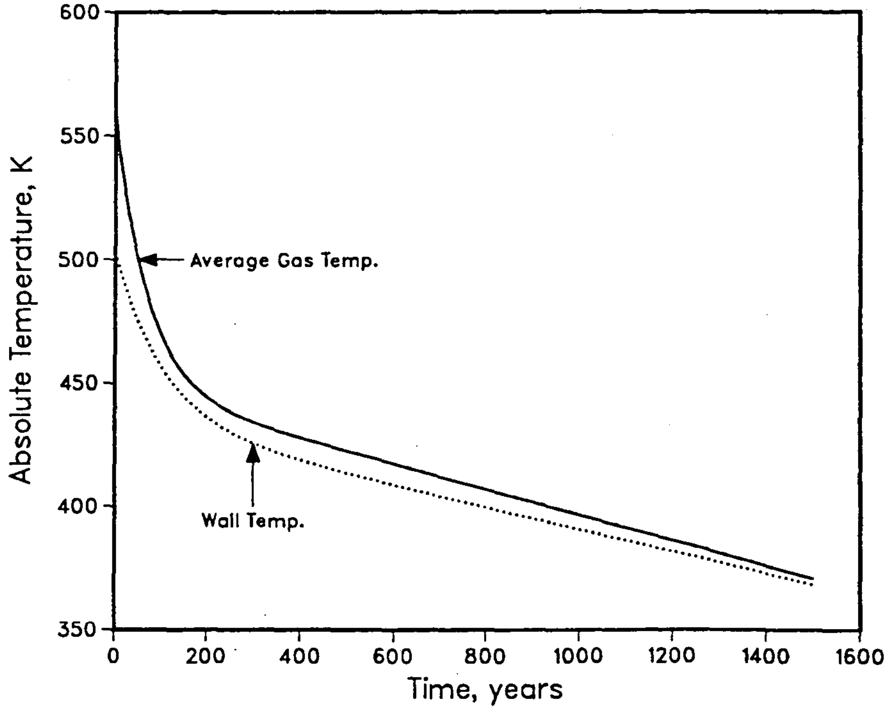

Figure 3. Waste Package Gas and Wall Temperature

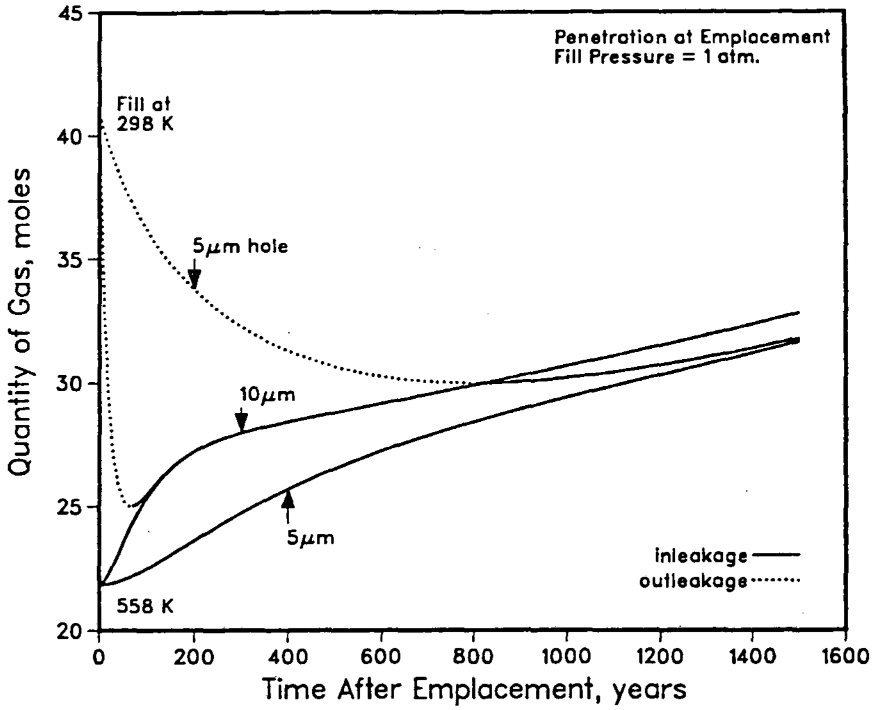

Figure 4. Quantity of Gas in a Container, Penetration at Emplacement

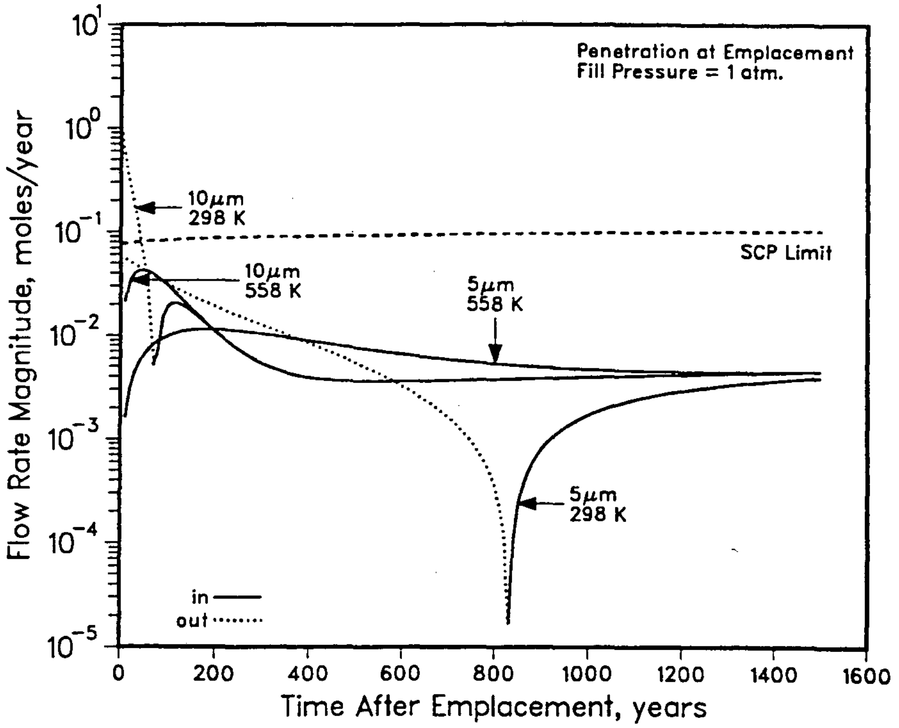

Figure 5. Gas Flow Rate Through a Penetration as Functions of Time, Aperture Size and Fill Conditions, Failure at Emplacement

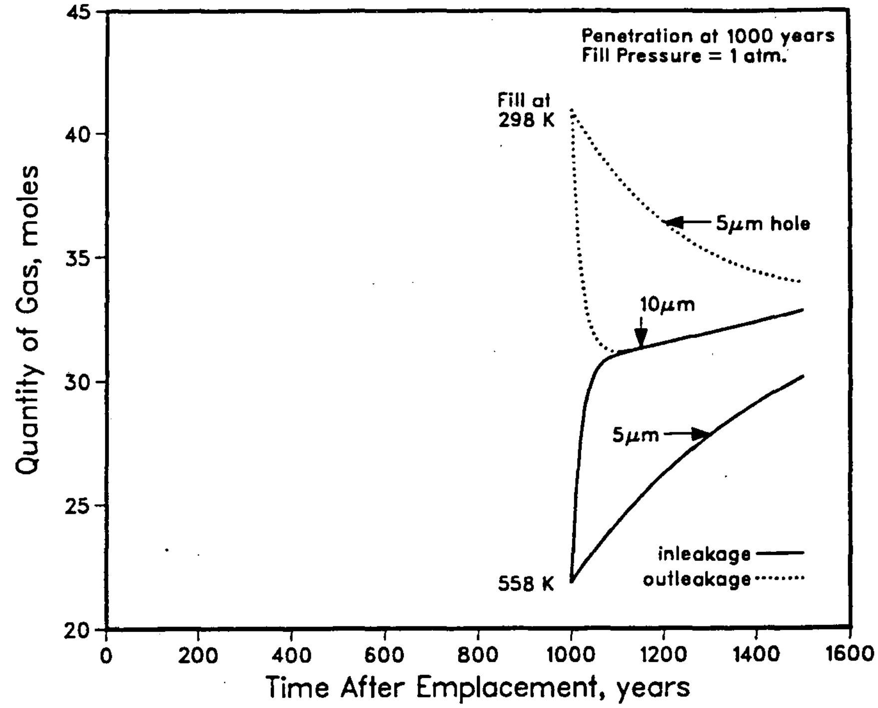

Figure 6. Quantity of Gas in a Container, Penetration at 1000 years

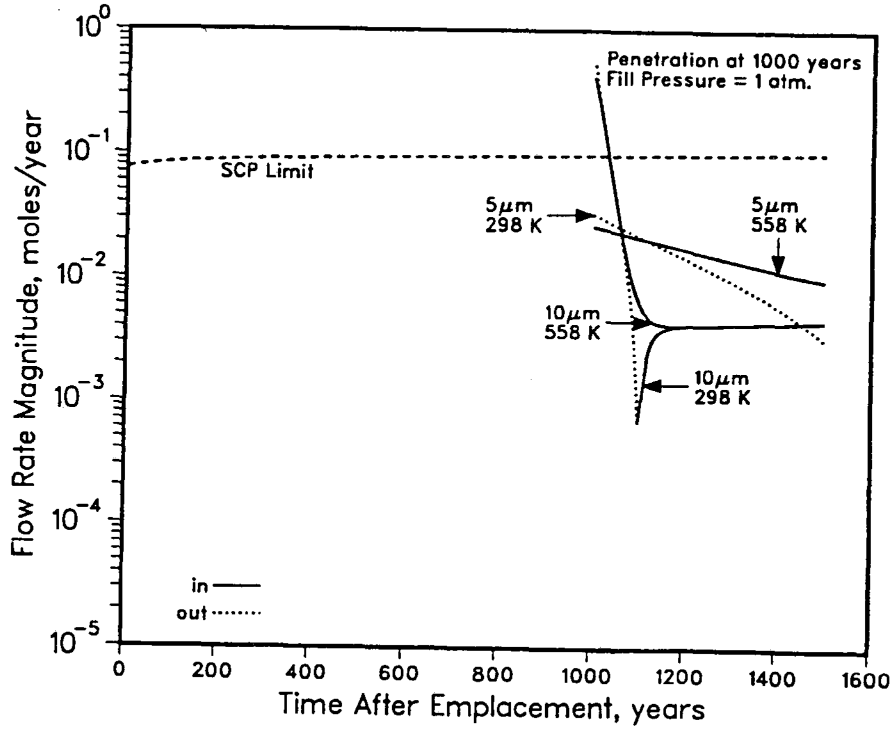

Figure 7. Gas Flow Rate Through a Penetration as Functions of Time, Aperture Size and Fill Conditions, Failure at 1000 years

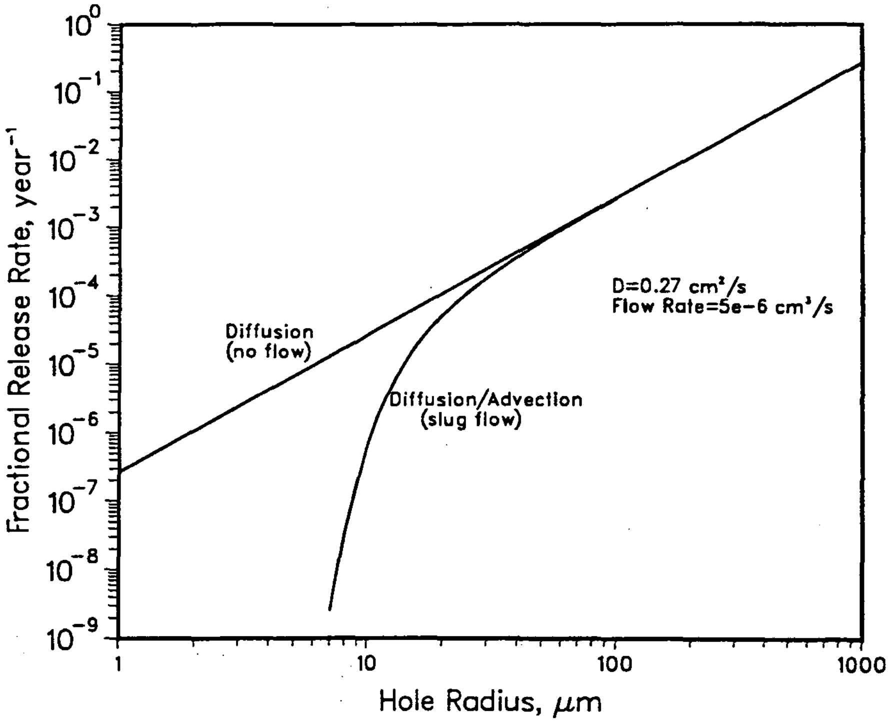

Figure 8. Fractional Release Rate with Slug Flow and Binary Diffusion

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UNIVERSITY OF CALIFORNIA
INFORMATION RESOURCES DEPARTMENT
BERKELEY, CALIFORNIA 94720