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**Release and Transport of Gaseous C-14 from
a Nuclear Waste Repository in an Unsaturated Medium**

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RELEASE AND TRANSPORT OF GASEOUS C-14 FROM A NUCLEAR WASTE REPOSITORY IN AN UNSATURATED MEDIUM

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ABSTRACT

The potential nuclear waste repository at Yucca Mountain is to be in partially saturated rock. Released radioactive gases such as $^{14}\text{CO}_2$ could have a direct pathway to the biosphere. We study the release of ^{14}C released as $^{14}\text{CO}_2$ from partly failed nuclear waste containers by analyzing the flow of gas into and out of a container. We analyze the transport of released $^{14}\text{CO}_2$ in an unsaturated, fractured, porous medium with gas-phase advection and dispersion. We calculate the inhalation dose to a maximally exposed individual above ground, based on some conservative assumptions about release from containers. For the assumed parameter values, a simple atmospheric diffusion model gives very small doses when compared to background radiation doses.

INTRODUCTION

Waste containers to be placed at the potential nuclear waste repository at Yucca Mountain will be filled with 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 $^{14}\text{CO}_2$. Eventually the waste cools, and together with the loss of inert gas through the penetration, the internal gas pressure drops. The pressure inside the container will eventually fall below atmospheric pressure. As the waste continues to cool, air leaking in can volatilize additional ^{14}C and oxidize the fuel.

We analyze this gas flow into and out of a nuclear waste container, through holes of specified timing and sizes and use these results to predict the release rate of $^{14}\text{CO}_2$.

We analyze the far-field transport of released $^{14}\text{CO}_2$ in an unsaturated, fractured, porous medium with gas-phase advection and dispersion. Gases released into a partially saturated, fractured rock tend to move in fractures, but some species are retarded by absorption in vadose water inside the rock matrix. Strong convective flow of gas is expected during the thermal phase of repository operation, carrying $^{14}\text{CO}_2$ rapidly toward the ground surface.

RELEASE OF C-14 FROM WASTE CONTAINERS

Gas Flow

The equations for steady flow of an ideal gas in a tube (Figure 1) of length ℓ and radius r , $\ell \gg r$, at constant temperature are [1]

$$\text{viscous } Q = \frac{\pi r^4 (P^2 - P_0^2)}{16\mu\ell RT_w} \quad (1a) \quad \text{or} \quad \text{molecular } Q = \frac{8\pi r^3 (p - p_0)}{3\ell\sqrt{2\pi m RT_w}} \quad (1b)$$

where Q is the molar flow rate of gas, T_w is the the container wall temperature, P and P_0 are the gas pressures inside and outside the container, m is the gas molecular weight, p and p_0 are the partial pressures of a gaseous species, and μ is the absolute viscosity of the gas. Because the temperature in the repository changes slowly, over hundreds of years, the steady-state gas flow law is applicable.

For $r \gg 1 \mu\text{m}$, the Knudsen number is $\ll 1$, and viscous flow applies [2]. We treat multiple penetrations in the container wall as a single hole of equivalent cross-sectional area. If molecular flow applies in multiple

holes individually, the total amount of gas flowing through the holes is small, and we conservatively assume viscous flow in a single equivalent-area hole. For holes that might result from pitting corrosion, the release of volatile species would be an impulse release of some of the inventory, with subsequent release controlled by diffusion. For holes greater than one μm , as well as cracks, advective transport is important and the Poiseuille equation (1a) can be used to establish the gas flow rates. In this study we consider the initial impulse release, advection, and diffusion.

In a waste container containing n moles of gas in a gas volume V , (1a) can be transformed into a differential equation in terms of n

$$\frac{dn(t)}{dt} = -\frac{\pi r^4 [(nRT/V)^2 - P_0^2]}{16\mu\ell RT_w} \quad (2)$$

with the initial conditions given by the ideal gas law

$$n(0) = \frac{P(0)V}{RT(0)} \quad (3)$$

The average gas temperature \bar{T} , and T_w the wall temperature, are known functions of time [3], and μ is an empirical function of T_w . The time for the waste to heat to its maximum temperature after emplacement is neglected.

Transport of a Tracer Gas Through a Hole

Viscous flow neglects any diffusive release of gaseous species. As fill gas leaks out through penetrations, gaseous species will be released by advection and diffusion, both in the same direction. At later times, as the waste package cools, air begins to flow into the waste container, and gaseous species will be released by diffusion against air inflow.

When advection and diffusion are in the same direction, advective transport of gaseous radionuclides out of the container is the dominant release process. When advection is inward and diffusion is outward, we have counter-diffusion, thus the diffusive transport of gaseous radionuclides is the only release mechanism.

We analyze counter-diffusion release through the equivalent tubular penetration, with quasi-steady-state flow and diffusion. The geometry is the same as Figure 1. Assuming slug flow and constant-concentration boundary conditions at the tube ends, the steady-state mass-balance equation for the radionuclide concentration c in the flow tube is

$$\bar{v} \frac{dc}{dz} = D \frac{d^2c}{dz^2}, \quad 0 \leq z \leq \ell \quad (4)$$

with z the direction of flow along the tube, and the boundary conditions

$$c(0) = c_0 \quad (5) \quad c(\ell) = 0 \quad (6)$$

Here c_0 is a constant concentration of the 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 conditions (5) and (6) ensure the maximum amount for diffusion, a conservative estimate.

The mass transfer rate \dot{m} is

$$\dot{m} = \pi r^2 \left(\bar{v}c - D \frac{dc}{dz} \right) \quad (7)$$

By solving (4)-(6) for c we obtain the fractional release rate f in terms of the mass transfer rate \dot{m}

$$f = \dot{m}/c_0V = \begin{cases} \pi r^2 \bar{v} \frac{e^{\bar{v}\ell/D}}{V(e^{\bar{v}\ell/D} - 1)}, & \bar{v} \neq 0; \\ \pi r^2 D/V\ell, & \bar{v} = 0 \end{cases} \quad (8)$$

This release rate is used to obtain the input to the far-field transport calculation.

FAR-FIELD TRANSPORT OF C-14

We assume that ^{14}C is released from nuclear waste as $^{14}\text{CO}_2(\text{g})$. Because CO_2 dissolves readily in water, we expect much of the ^{14}C to be retarded by absorption into vadose water. We neglect precipitation into solid phases. However, we utilize the capacity of calcite to maintain a constant pH in vadose water.

We define a gas-liquid distribution coefficient for equilibrium concentrations

$$K_D = \frac{[\text{CO}_2(\text{aq})] + [\text{H}_2\text{CO}_3(\text{aq})] + [\text{HCO}_3^-] + [\text{CO}_3^{2-}]}{[\text{CO}_2(\text{g})]} \quad (9)$$

and note that K_D is both temperature and pH dependent. We use this relationship to approximate the ratio of ^{14}C in liquid to that in the gas phase at the gas/liquid interface.

First, we consider a single vertical planar fracture filled with gas, adjacent to a porous medium. We neglect dispersion in the fracture and assume that the gas is well-mixed over the width of the fracture so that the concentration is uniform in the fracture. We also assume uniformity in the x -direction for concentrations in the fracture and rock matrix, and we assume a constant, upward gas velocity in the fracture.

We define a Peclet number as

$$\text{Pe} \equiv \frac{bv_g}{\epsilon_g D_g + \epsilon_l D_l K_D} \quad (10)$$

where b is the fracture half width [L],

D_l is the liquid-phase diffusion coefficient [L^2/t],

ϵ_g is the gas-filled porosity, and

D_g is the pore-gas diffusion coefficient [L^2/t],

v_g is the gas velocity in the fracture [L/t],

ϵ_l is the liquid-filled porosity.

We conclude that if the Peclet number is much less than unity, the liquid concentrations in the rock matrix between fractures will be well equilibrated with gas concentrations in the fractures. For details of this analysis see Light *et al.* [4].

If there is equilibrium between ^{14}C in the fractures and in all rock-matrix pore liquid at a given z , we can treat the whole as an equivalent porous medium without regard to fracture details. Assuming local equilibrium, we replace $C_l(\mathbf{r}, t)$ with $K_D C_g(\mathbf{r}, t)$ in the general porous-medium governing equations and assume constant coefficients, yielding

$$\left(\frac{\partial}{\partial t} + \lambda\right) C_g(\mathbf{r}, t) + \mathbf{v} \cdot \nabla C_g(\mathbf{r}, t) - D \nabla^2 C_g(\mathbf{r}, t) = \frac{f(\mathbf{r}, t)}{\epsilon_g + \epsilon_l K_D} \quad (11)$$

where

$$\mathbf{v} = \frac{\mathbf{q}_g + \mathbf{q}_l K_D}{\epsilon_g + \epsilon_l K_D}, \quad D = \frac{\epsilon_g D_g + \epsilon_l D_l K_D}{\epsilon_g + \epsilon_l K_D} \quad (12)$$

Here \mathbf{q}_g is the gas Darcy velocity, \mathbf{q}_l is the liquid Darcy velocity, and $f(\mathbf{r}, t)$ is a source term. The scalar diffusion coefficients D_g and D_l have been used in place of dispersion tensors.

We solve (11) for various source terms, boundary conditions and initial conditions. All mathematical details are given in our technical report [5]. We assume an infinite domain with the concentration equal to zero initially and vanishing at infinity for all time. This assumption does not allow for a boundary condition at the ground surface. In reality, the ^{14}C gas concentration at the ground surface is near zero because of atmospheric dispersion. Therefore, the ^{14}C flux may be somewhat greater than predicted here due to the increased concentration gradient at the ground surface.

We consider an infinite plane source located in the $z = 0$ plane, releasing an amount I_f over the repository area A . The solution for an impulse release at time t_f is

$$C_g(z, t) = \frac{I_f}{A} \frac{\exp\left[-\frac{(z-v(t-t_f))^2}{4D(t-t_f)} - \lambda(t-t_f)\right]}{(\epsilon_g + \epsilon_l K_D)[4\pi D(t-t_f)]^{1/2}}, \quad t > 0 \quad (13)$$

The flux from an infinite plane source with an impulse release of strength I_f/A is

$$\Phi(z, t) = \frac{I_f}{A} \frac{\exp \left[-\frac{(z-v(t-t_f))^2}{4D(t-t_f)} - \lambda(t-t_f) \right]}{[4\pi D(t-t_f)]^{1/2}} \left[\frac{z+v(t-t_f)}{2(t-t_f)} \right], \quad t > 0 \quad (14)$$

We deal with the release rate from the partly failed container by superpositioning impulse releases.

The above-ground $^{14}\text{CO}_2$ concentration c [Ci/L^3 or Bq/L^3] is estimated by a method after Schiager [6] for estimating exposures from a diffuse underground source

$$c = \frac{\Phi L}{\sigma_z U} \quad (15)$$

where L is the lateral extent of the repository [L], U is the mean wind speed [L/t], and σ_z is the vertical standard deviation of the assumed Gaussian distribution of concentration [L].

To convert above-ground $^{14}\text{CO}_2$ concentration to dose, we use the dose factor from a NCRP method [7].

$$10^8 \frac{\text{mrem/a}}{\text{Ci}/\text{m}^3} \quad (16)$$

ILLUSTRATIVE RESULTS

C-14 Release from a Partly Failed Container

We integrate (2) 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 obtained from *Site Characterization Plan*, [3; p. 7-41] and extrapolated in time. We use a container thickness, ℓ , of 0.01 m. 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 ($5 \mu\text{m}$) and $10 \mu\text{m}$, and two temperatures at time of sealing, 298 K and 558 K. The fill pressure is assumed to be 0.1 MPa, and the hole is assumed to occur at 300 years after the waste is buried. Figure 2 shows the number of moles of gas inside the waste container as a function of time, and the flow rate through the hole is the slope of the lines in Figure 2. The dotted lines refer to outleakage and the solid lines refer to inleakage. For a $5\text{-}\mu\text{m}$ hole, and a seal temperature of 298 K, argon slowly leaks out until about 1100 years, after which leaking and repository cooling cause the internal pressure to fall below atmospheric, and air leaks in. For a $5\text{-}\mu\text{m}$ hole, 24% of the argon flows out, whereas for a $10\text{-}\mu\text{m}$ hole, 33% flows out. For a $10\text{-}\mu\text{m}$ hole, the internal pressure rapidly falls to atmospheric, and inleakage begins at about 400 years. For a 558 K seal temperature, the maximum gas temperature in waste packages, there is no thermally generated buildup of pressure and thus no gradient for outleakage. Here a penetration results only in inleakage of air because the internal pressure is always less than atmospheric.

Figure 2 can be used to estimate release rates. For a 298 K seal temperature and a $5\text{-}\mu\text{m}$ hole, about one-fourth of the gas and volatilized radionuclides leaks from the container. For $10\text{-}\mu\text{m}$ holes or larger, almost half of the gas leaks out before air inleakage begins. If we assume that $^{14}\text{CO}_2$ was released from the fuel cladding surface at the time of maximum temperature and it leaks out congruently with argon, then we can compute a $^{14}\text{CO}_2$ molar flow rate assuming no further volatilization of CO_2 during this period.

Assuming that heating of the waste package volatilizes 10 percent of the ^{14}C inventory [8], primarily from cladding surfaces, a $5\text{-}\mu\text{m}$ hole and 298 K seal temperature result in an initial argon leak rate of 0.06 mole/a and a ^{14}C initial fractional leak rate of $2 \times 10^{-5}/\text{a}$. For a seal temperature of 558 K or higher, no radioactive material will be released if counter-diffusion is neglected.

To calculate $^{14}\text{CO}_2$ release by combined advection and diffusion we combine the solution to the viscous flow model, (2) and (3), and the solution for diffusive release, (8). At each time-step we find the average

viscous-flow velocity \bar{v} and calculate the release rate using (8). Using a mass balance at each time step, we calculate c_0 , the ^{14}C concentration inside the container in (5). Thus the depletion of ^{14}C in the container is accounted for.

Because counter-flow is a resistance to diffusion, assuming $\bar{v} = 0$ will give an upper bound to the counter-diffusion release rate. For a 5- μm hole and 10 percent ^{14}C volatilized, the diffusive fractional release rate is $7 \times 10^{-7} \text{ year}^{-1}$ based on total ^{14}C inventory. For holes larger than 30- μm the flow is slow and diffusion is the dominant release mode.

For extremely small holes, molecular flow applies. To show the relative magnitudes of viscous flow and molecular flow, we calculate the flow rate of argon at 500 K through various aperture sizes, under identical pressure gradients ($P = 0.19 \text{ MPa}$, $P_0 = 0.1 \text{ MPa}$). The results are shown in Table I. Values in italics are for the flow regime that actually occurs. Other values are hypothetical. Table I shows that molecular flow occurs in the smaller holes, but the actual flow rate and quantity of gas moving through the such small penetrations is miniscule and unimportant. For the 1- μm hole in Table I, the mean free path is one tenth the hole radius, therefore viscous flow occurs. The actual penetration sizes that result in the overall single penetration considered here may be individually small enough for Knudsen flow to occur. Assuming viscous flow through these larger equivalent single-hole penetrations is conservative.

If the actual penetrations are few in number and are near the size of the equivalent single holes considered here, the actual flow regime will be viscous, as has been analyzed. Even if the distribution of hole sizes includes many small holes in which Knudsen flow occurs, viscous flow through even a few larger holes is likely to dominate the leak rate. This is illustrated by the calculated results in Table I.

Table I. Comparison of Molecular and Viscous Flows of Argon for the Same Pressure Gradient, $T = 500 \text{ K}$, $P = 1 \text{ MPa}$, $P_0 = 0.1 \text{ MPa}$, $\ell = 0.01 \text{ m}$

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

Italics are actual flow; others are hypothetical.

We now calculate ^{14}C release rates from a single waste container, using
 Container gas volume, $V = 1.22 \text{ m}^3$ External gas pressure, $P_0 = 0.1 \text{ MPa}$
 Internal gas fill pressure, $P = 0.1 \text{ MPa}$ Internal gas seal temperature, 298 K
 Initial ^{14}C inventory 3.1 Ci ($1.1 \times 10^{11} \text{ Bq}$) per package Volatile ^{14}C fraction = 10% (assumed, after [8])
 Diffusion coefficient of CO_2 in air = $0.27 \times 10^{-4} \text{ m}^2/\text{s}$ [9]

We calculate ^{14}C release rates for holes of one to 300- μm occurring at emplacement and 300 years. These release rates consist of an initial pulse and a longer term release. For holes occurring at 300 years, we calculate an initial release of 0.097 Ci/package ($3.6 \times 10^9 \text{ Bq/package}$). For the long-term release, a time-dependent release rate of ^{14}C can be obtained by using the maximum release rate for various hole sizes at each time. This release rate is used in far-field transport calculations.

Far-Field Transport Results

For calculations using (13) and (14), we use b , fracture half width, 10^{-5} m [10]; $K_D = 3$, for pH and 50°C ; D_g pore-gas diffusion coefficient, 1.6×10^{-6} m²/s; D_l liquid-phase diffusion coefficient, 9.5×10^{-11} m²/s; ϵ_g gas-filled porosity, 0.02; and ϵ_l liquid-filled porosity, 0.08. We use a gas Darcy velocity calculated by Tsang and Pruess [11] of 0.04 m/y or a fracture pore velocity of 22 m/y. We assume that $v_g \gg v_l$ and set $q_l = 0$.

For release of $^{14}\text{CO}_2$ from waste in the proposed repository at Yucca Mountain we calculate a Peclet number of approximately 2×10^{-4} . This indicates that ^{14}C in pore liquid at fracture surfaces will be retarded in accord with the local-equilibria assumption used in the equivalent-porous-medium model.

To estimate above-ground $^{14}\text{CO}_2$ concentrations, the following values are used in (15): $L = 3000$ m, $\sigma_z = 20$ m. An atmospheric turbulence class of F, moderately stable conditions, has been assumed [12]. Currently no site-specific wind speeds are available for Yucca Mountain. In the *Site Characterization Plan*, surface winds at Yucca Mountain have been estimated using historical wind data from four meteorological towers at Yucca Flat at the Nevada Test Site [3; p. 5-21]. Wind speeds are generally less than 5.4 m/s and 13.1 % of the time it is calm. Therefore we conservatively use $U = 1$ m/s.

Figure 3 shows $^{14}\text{CO}_2$ annual releases at a point 350 m above the repository if a hole occurs in all containers in the repository at 0 or 300 years, with gas Darcy velocity as a parameter. Water saturation is taken to be 0.8. The y -axis on the right shows the dose to a maximally exposed individual from air immersion, ground surface exposure and inhalation, calculated using the method discussed above in (11) to (14). The dose to that individual is very low, approximately 0.1% of background, for the peak release at a gas Darcy velocity of 0.4 m/a, an order of magnitude higher than the calculated gas Darcy velocity [11]. This is from some extreme assumptions about container integrity.

Figure 4 shows cumulative releases of ^{14}C as a function of time of hole occurrence and gas Darcy velocity. The difference in steady-state cumulative releases due to different gas Darcy velocity is attributable to decay. The difference in cumulative amounts of the hole occurring at 0 year and 300 years is because we use a slightly different release rate for far-field transport. It is interesting to note that if the gas Darcy velocity is as high as 0.4 m/a, then practically all of the ^{14}C released reaches the ground surface within several hundred years. If the gas Darcy velocity is 0.04 m/a, then it will take several thousand years, but well short of one half-life of ^{14}C , for all the released ^{14}C to reach the ground surface.

CONCLUSIONS

The release of ^{14}C as $^{14}\text{CO}_2$ from partly failed spent fuel containers has been analyzed by the flow of gases into and out of the containers. This flow of gases is driven by pressure differences, which are in turn caused by heating by the spent fuel. The transport of released $^{14}\text{CO}_2$ has been analyzed by transport in equivalent porous medium. Our calculations, using the particular data set, show that essentially all the released ^{14}C will reach the ground surface in less than one half-life of ^{14}C . However, the quantities of ^{14}C reaching the ground surface are so small that even if all containers fail at emplacement and conservative dose factors are used, the resultant inhalation dose to the maximally exposed individual is about 0.01% of natural background radiation.

These conclusions may be incorrect, however, if during site characterization it is discovered that fracture zones form a direct, connected pathway for gas transport from repository horizon to the ground surface. In such a case the equivalent porous medium model we use is not valid.

We also need information about the container material and design so that the mode of container failure can be factored into our analysis.

ACKNOWLEDGEMENT

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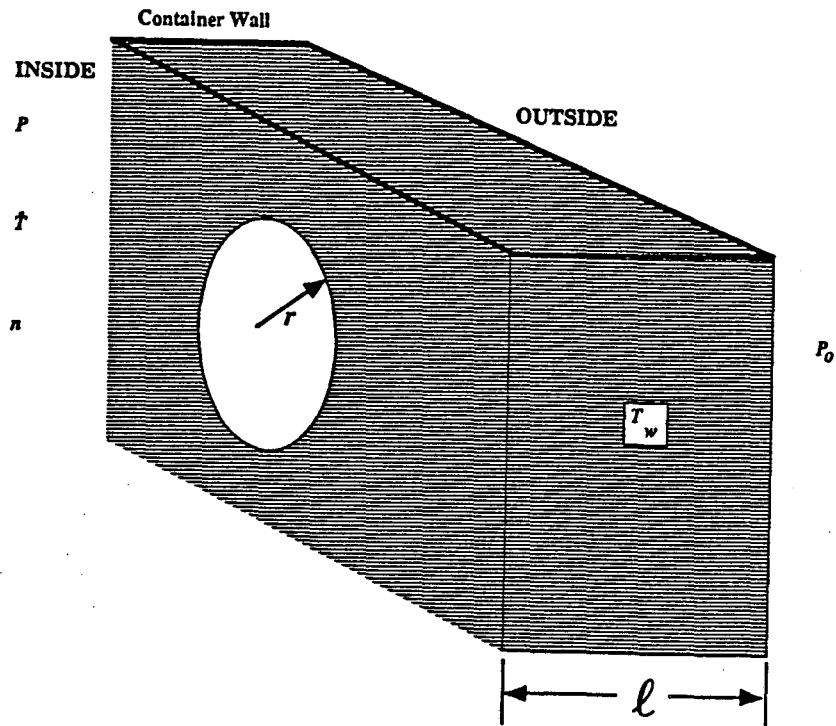


Figure 1. Penetration in a Container Wall

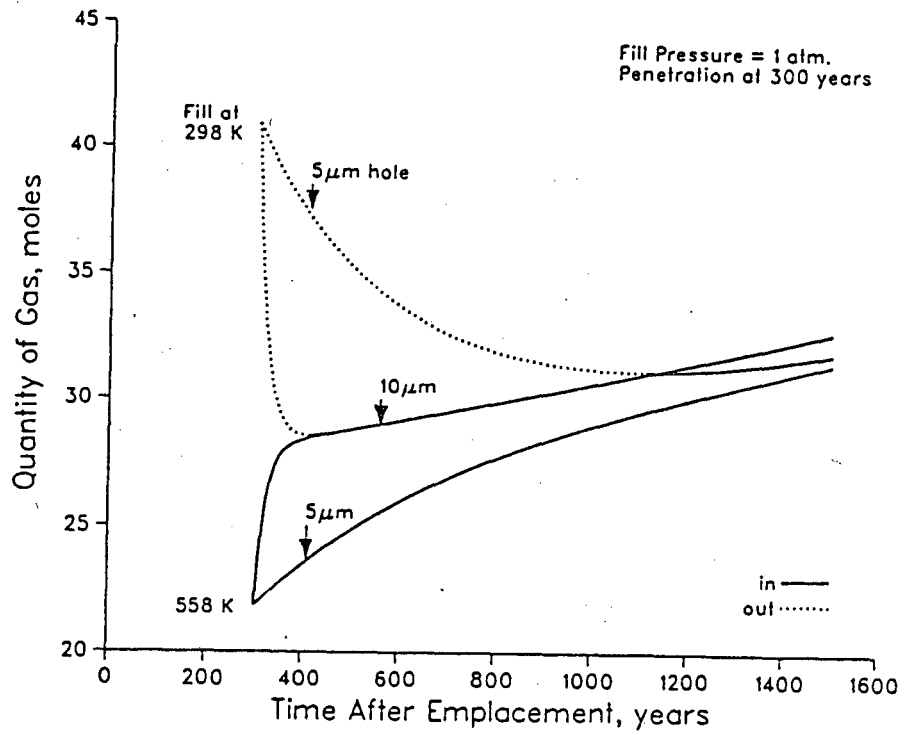


Figure 2. Quantity of Gas in a Container, Hole at 300 Years

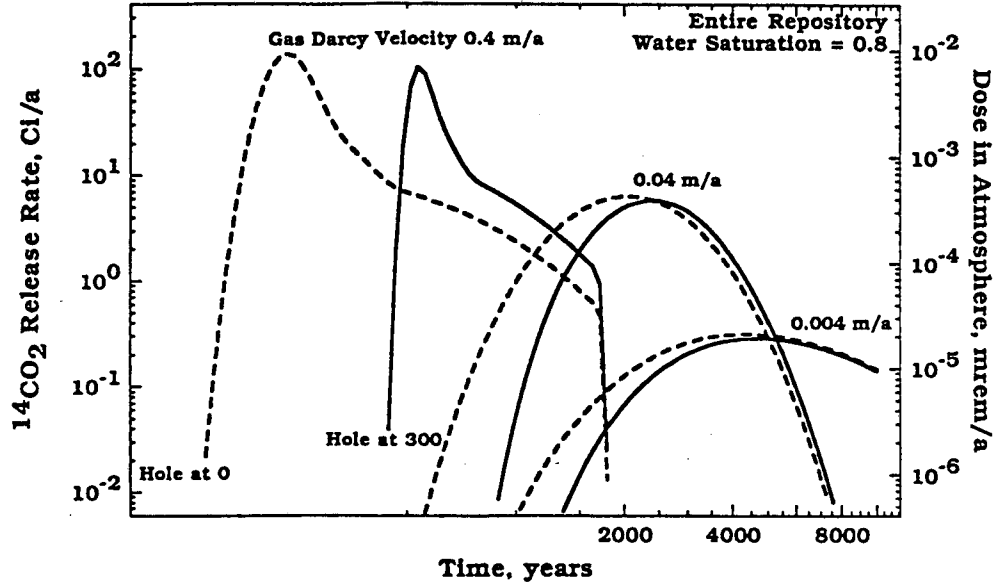


Figure 3. Annual Release Rate and Dose of C-14 at Ground Surface, Hole at 0 or 300 Years

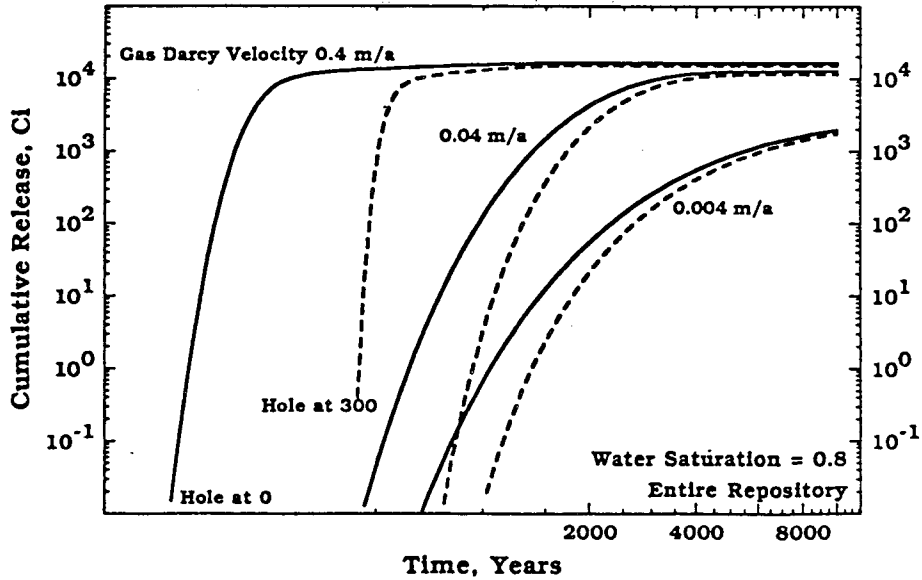


Figure 4. Cumulative Releases of C-14 at Ground Surface, Hole at 0 or 300 Years

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