TRANSIENT AND STEADY-STATE RADIONUCLIDE TRANSPORT THROUGH PENETRATIONS IN NUCLEAR WASTE CONTAINERS

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ABSTRACT

In this paper we analyze the transport of radionuclides through penetrations in nuclear waste containers. Penetrations may result from corrosion or cracks and may occur in the original container material, in degraded or corroded material, or in deposits of corrosion products. We do not consider how these penetrations occur or the characteristics of expected penetrations in waste containers. We are concerned only with the analytical formulation and solutions of equations to predict rates of mass transfer through penetrations of specified size and geometry. Expressions for the diffusive mass transfer rates through apertures are presented. We present numerical illustrations for steady-state mass-transfer rates through a circular hole, including concentration isopleths. The results are extended to multiple holes, including a criterion for hole spacing wherein superposition of single-hole solutions can be used. Results illustrated for holes in thin-walled containers show that significant mass transfer can occur even if a small fraction of the container area is perforated. We also illustrate the case of holes facing a water gap, instead of being in intimate contact with porous rock. In this case the radionuclide flux from many small holes approaches that from a bare waste cylinder.

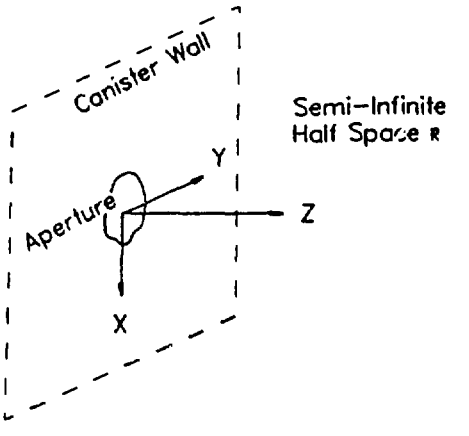
INTRODUCTION

Studying mass transfer through container penetrations is relevant to demonstrating compliance with the U. S. Nuclear Regulatory Commission's performance objectives [1] for the waste-package subsystem that call for substantially complete containment within the waste package for 300 to 1000 years and require 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. In this paper we analyze the transport of radionuclides through penetrations in waste containers. Penetrations may result from corrosion or cracks and may occur in the original container material, in degraded or corroded material, or in deposits of corrosion products. We do not consider how these penetrations occur or the characteristics of expected penetrations in waste containers. We are concerned only with the analytical formulation and solutions of equations to predict rates of mass transfer through penetrations of specified size and geometry. Expressions for the diffusive mass transfer rates through apertures and numerical illustrations are presented.

MASS TRANSFER EQUATIONS

The following assumptions are made. The transport of radionuclides from an opening into the surrounding porous rock is by diffusion only. This space consists of a water-saturated medium, sufficiently fine grained compared with the aperture dimensions, to validate the diffusion mechanism into a continuum. The radionuclide in the waste form supplies a

uniform concentration over an assumed time-invariant cross section of the aperture. The spatial extent of the concentration field away from the opening is small, as shown below, so that the canister wall can be treated as an infinite plane. Figure 1 shows the situation studied and the coordinate system.



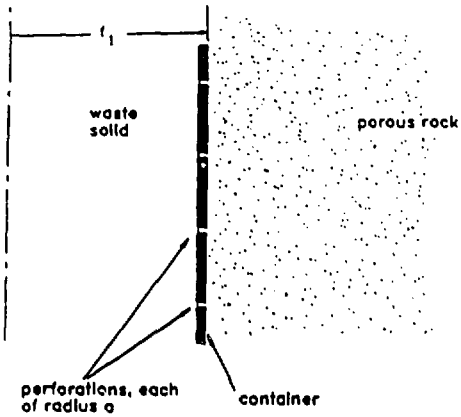


Figure 1a. The Coordinate System. Figure 1b. Situation Studied

The governing equations for the radionculide concentration N(X,Y,Z,t) are

$$\frac{\partial N}{\partial t} = \frac{D_f}{K} \nabla^2 N - \lambda N, \text{ valid in } R,\ t > 0 \tag{1}$$

$$N = 0, \text{ valid in } R,\ t = 0 \tag{2}$$

$$V \frac{dN_w}{dt} = - D_f \epsilon_o \int_{S_a} \text{grad } N d\sigma - \lambda V N_w,$$
$$\text{valid on aperture surface } S_a,\ t \geq 0 \tag{3}$$

$$- D_f \epsilon_o \text{ grad } N = 0 \quad \text{valid on unimpaired container surface, } t \geq 0. \tag{4}$$

N is a bounded function in R, for $t > 0$. D_f and K are the diffusion and retardation coefficients of the radionuclide and λ its decay constant. ϵ_o is the porosity of the water-saturated medium. N and N_w are the radionuclide concentrations in the half space and waste form respectively. V is the waste form volume. S_a is the aperture surface area and a is a characteristic dimension of the aperture.

Equation (1) describes the radionuclide balance due to diffusion, decay and storage in R. Equation (2) states that, prior to the aperture opening, the specie concentration in R is zero. Equation (3) describes the rate of change of the radionuclide in the waste form due to diffusive mass transport through the opening and due to decay. It furnishes the nuclide concentration on the aperture surface, since $N = N_w$ there. We assumed $dN_w/dt=0$, or a time-invariant nuclide concentration at the waste surface. Equation (4) indicates the absence of diffusive mass transport through the impermeable part of the canister surface. The purpose of the following analysis is the determination of the time-dependent mass flux

through the aperture and the associated concentration field at steady state.

The solution to the governing equations presented above [2] is

$$\dot{m}(t) = D_f \epsilon_o (n_w e^{-\lambda t}) aS(t) \tag{5}$$

where $\dot{m}(t)$ is the radionuclide transfer rate from an aperture with a characteristic dimension of a, S(t) is the shape factor for the aperture, and $n_w = N_w/e^{-\lambda t}$. As indicated in eq. (5), the shape factor is a function of time, so that the transient mass flux is a strong function of the shape of the hole. In reference 2, we provide a more detailed discussion of the time-dependent shape factor, for regular and irregular-shaped holes. For circular holes

$$S(\theta) = 4\left(1 + \frac{4}{(4\pi^3\theta)^{1/2}}\right), \quad \theta \to \infty \tag{6}$$

where

$\theta = tD_f/Ka^2$, dimensionless time

and

$$S(\theta) = A/\sqrt{\pi\theta}, \quad \theta \to 0 \tag{7}$$

Using eq. (5), we can obtain the dimensionless, steady-state radionuclide concentation $c^s(r,z)$ near a hole at $r^2 = X^2/a^2 + Y^2/a^2$ and $z=Z/a$,

$$c^s(r,z) = \frac{2}{\pi} \sin^{-1}\left(\frac{2}{\sqrt{z^2 + (1+r)^2} + \sqrt{z^2 + (1-r)^2}}\right). \tag{8}$$

This result was also obtained by Rae [3].

NUMERICAL ILLUSTRATIONS

Diffusion Through Single Holes

We now provide numerical and graphical illustrations of the application of equation (8). In all examples, the aperture or apertures are circular and on cylindrical waste containers. Figure 2 shows the steady-state isopleths around a single aperture with radius a, calculated using eq. (8). At the position r=0 and z=0.5, the steady-state concentration is less than 70 percent of the hole surface concentration, while at r=0 and z=12.7 the concentration has dropped to 5 percent. Also shown in dashed lines in Figure 2 are the diffusion paths, normal to the concentration gradients.

The dimensional diffusive flux is

$$j(r,a) = \frac{2D_f \epsilon_o n_w}{\pi a\sqrt{1 - r^2}}, \quad 0 < r \leq 1 \tag{9}$$

When the normalized distance along the hole is unity, the diffusive mass flux j(r, a) becomes infinite, consistent with the observation from Figure 2 that the diffusive mass transfer is most intense near the hole edge.

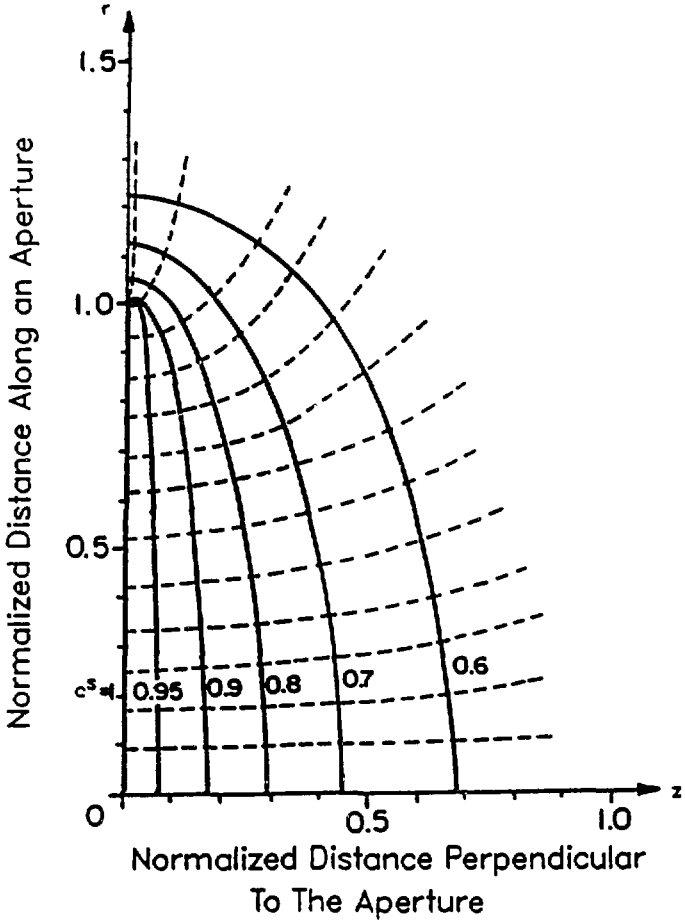


Figure 2. Surfaces of Non-Dimensional Concentrations and Diffusional Paths

The steady-state dimensional mass release rate through a circular hole of radius a is

$$\dot{m}^s(a) = \int_0^a 2\pi r j(r,a)dr = 4D_f \varepsilon_o a n_w \qquad (10)$$

Therefore, the mass release rate through a hole increases linearly with the hole radius, magnitude of the diffusion coefficient, surface concentration, and porosity of the rock. This result is expected to be valid in porous media of small porosity, or for a hole size much larger than the grain size of the porous medium adjacent to the container.

We now extend the results of the single, circular hole analysis to multiple apertures on a container. It is assumed that all apertures are circular and of an identical size.

Diffusion Through Multiple Apertures Into a Porous Medium

If multiple apertures are present on a waste cylinder, and if one seeks to predict the total mass transfer rate from all holes, the total mass transfer rate is the sum of contributions from all holes. For this sum to be obtained from the single-hole equations, one must first determine if the concentration fields from adjacent holes overlap, causing the concentration fields to interfere with the prescribed concentration boundary condition at the aperture. In other words, how far must the holes be separated for the summation to be valid? From eq. (8), it is seen that at z=0, r=10, then c^S=0.064, and the concentration has fallen to negligible levels about 10 hole radii away, so that the summation rule might apply. For example, on a cylindrical container, with a radius of 0.15 m and a length of 2.46 m, there can be 235 identical 1-cm radius holes, or 23,000 0.1-cm holes, for this summation rule to be valid.

For holes sufficiently separated, the total mass transfer rate through ν holes of radius a would be

$$\dot{m}_a^S(\nu, a) = \nu \dot{m}^S(a) \tag{11}$$

Or from equation (10),

$$\dot{m}_t^S(\nu, a) = 4 \nu D_f \varepsilon_0 a N_w \tag{12}$$

It may be instructive to compare the total rate of release from holes on a container to the release rate from a bare cylinder of the waste form, both subject to diffusion only. The steady-state diffusive mass transfer rate from a waste form cylinder of radius f_1 and length ℓ in contact with porous rock is given by equation (7.5.2) of Chambre´ **et al.** [4],

$$\dot{m}_c^S(f_1, \ell) = 2\pi D_f \varepsilon_0 N_w \ell / \log(\ell / f_1) \tag{13}$$

where the end effects have been neglected.

To compare the two release rates for fixed waste-cylinder dimensions, define the following dimensionless mass transfer ratio:

$$\alpha(\nu, a) = \dot{m}_t^S(\nu, a) / \dot{m}_c^S(f_1, \ell) \tag{14}$$

The dimensionless mass transfer ratio is the total rate of mass transfer from apertures of radius a to the total mass transfer rate from a cylinder with no cladding. Using equations (12) and (13) one can rewrite equation (14) in the following form:

$$\alpha(\nu, a) = \frac{2a\nu}{\pi \ell} \log(\ell / f_1). \tag{15}$$

In Figure 3, $\alpha(\nu, a)$ is plotted as a function of ν for aperture radii of a=1 cm, 1 mm, and 0.5 mm, located on the surface of a waste form with f_1= 0.15 m, ℓ =2.46 m. Ratios of the total hole area to the total cylindrical surface (including holes), but excluding end surfaces, are indicated on each of the three lines of $\alpha(\nu, a)$. Figure 3 shows that the radionuclide transfer rate from a waste container with numerous small holes can actually exceed that from a bare waste cylinder of the same dimensions. On a bare waste form cylinder, the mass transfer rate would be uniform over the surface of the cylinder, but in a cylinder with numerous holes, the gradient at all hole edges is infinite, leading to more intense mass transfer. For a waste cylinder of f_1=0.15 m and ℓ = 2.46 m,

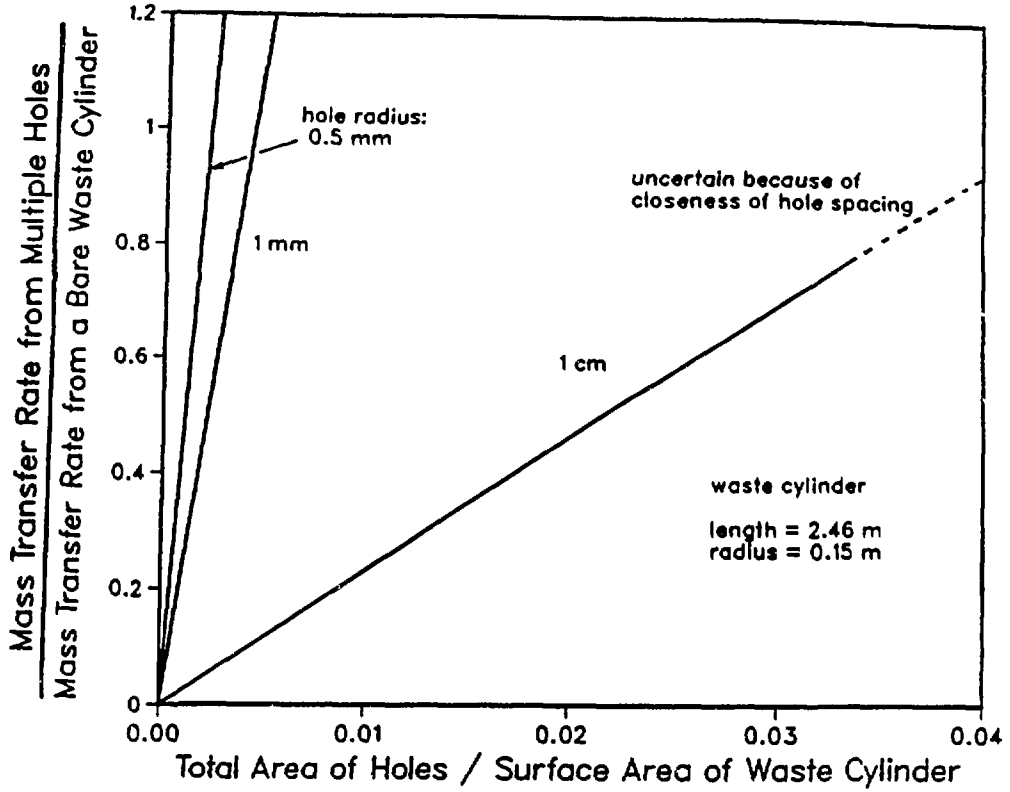


Figure 3. Radionuclide Transfer Rates from Multiple Apertures and a Bare Waste Cylinder

the mass transfer ratio of eq. (15) can be written as

$$\alpha(\nu, a) = 3.13 \times 10^{-3}\, \nu a \qquad (16)$$

If all holes are 1 mm and there are 3,200 holes, then $\alpha > 1$, and the mass transfer rate from a waste cylinder with holes exceeds that from a bare waste cylinder of equivalent dimensions. As the number of holes increases and the holes become closely spaced, the concentration fields from adjacent holes begin to overlap appreciably, and the superposition of single-hole solutions is no longer valid. This occurs at an area ratio of about 0.033 for holes of 1 cm radius.

Diffusion Through Multiple Apertures Into a Water Gap

In actual repository environments, the perforated container might not be in intimate contact with the surrounding porous media due to rock movement, creating a void space between the holes and the porous rock. A water gap might also occur if the holes are in fuel elements that have been overpacked without the matrix being filled. A water gap between the perforated container and the porous medium, as shown in Figure 4, can result in release rates different from those estimated

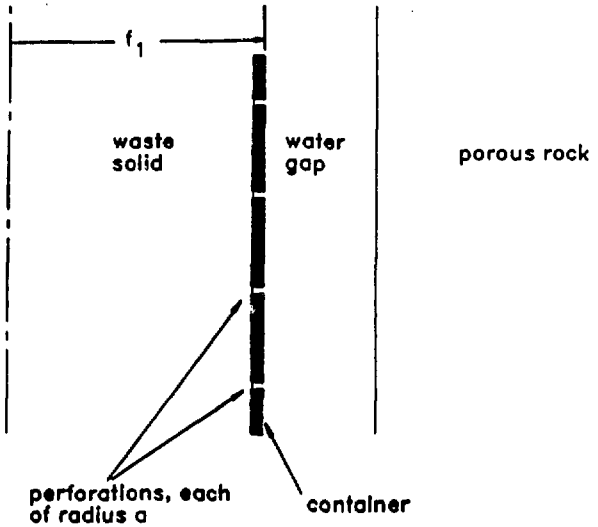


Figure 4. Geometry for a Water Gap Between Container and Porous Rock

above. To illustrate, we develop a simplified analysis by assuming no convection in the water gap.

The calculations above for a perforated container in contact with an infinite porous medium also apply to a container in contact with infinite water, of unit porosity, if convection is negligible and transport is entirely by diffusion. The analytical solutions presented earlier for steady state assumed zero concentration in water at an infinite distance in the Z direction from the container. If there is a finite far-field concentration N_∞ in ground water far from the waste, the analytical solutions can be used by redefining $c^s(r,z)$ and rewriting equation (8) as:

$$c^s(r,z) = \frac{N(r,z) - N_\infty}{N_w - N_\infty} = \frac{2}{\pi} \sin^{-1}\left(\frac{2}{\sqrt{z^2+(1+r)^2} + \sqrt{z^2+(1-r)^2}}\right) \qquad (17)$$

and equation (12) for the mass transfer rate $\dot{m}_t^s$ through ν apertures in a container surrounded by water, with no convection, becomes:

$$\dot{m}_t^s = 4\nu D_f a (N_w - N_\infty) \qquad (18)$$

From equation (17) we find that at normalized distances z = 10 and 100 the concentration differences are:

$$(N(0,z) - N_\infty) / (N_w - N_\infty) = 0.064 \text{ , for } z = 10$$
$$= 0.0064 \text{ , for } z = 100$$

Thus, to a good approximation, for a container with 1-mm holes a few centimeters or more of water are sufficient for the water to appear to the holes as an infinite diffusing medium.

From equation (17) it can be demonstrated that at a distance z from the container equal to or greater than ten hole radii, the concentration in the water is essentially constant over a plane normal to Z. Therefore, we expect that if the porous medium is positioned ten hole radii or more from the container it will be exposed to a concentration that is essentially uniform over its surface.

For these reasons, we will use equation (18), derived for an infinite

diffusing medium, as an approximation for the water gap of finite thickness, and we will adopt the concentration N_r at the porous-medium interface as the concentration N in equation (18). There results:

$$\dot{m}_t^s = 4 \nu D_f \, a \, (N_w - N_r) \tag{19}$$

The steady-state mass-transfer rate $\dot{m}_t^s$ is also given by applying equation (13), but with N_r as the concentration and f_1 as the radius at the inner surface of the medium of porosity ε_0:

$$\dot{m}_t^s = 2 \pi D_f \, \varepsilon_0 N_r \, \ell / \log(\ell / f_1) \tag{20}$$

Combining equations (19) and (20) to eliminate N_r, we obtain

$$\dot{m}_t^s = \frac{2\pi D_f \varepsilon_o N_w \ell}{\log(\ell/f_1) + \frac{\pi \varepsilon_o \ell}{2\nu a}} \tag{21}$$

Now, we seek an expression for the steady-state release through the water gap and porous medium if the waste container is not present. Because of the low diffusional resistance of a few centimeters of water continuum as compared with that of the surrounding low-porosity rock, we expect that the steady-state concentration N_r will be close to the concentration N_w at the waste surface. Assuming $N_r = N_w$ for no container, the steady-state mass-transfer rate is:

$$\dot{m}_t^s = 2 \pi D_f \, \varepsilon_0 N_w \, \ell / \log(\ell / f_1) \tag{22}$$

The ratio $\alpha(\nu, a)$ of the mass-transfer rates with and without a perforated container is then:

$$\alpha(\nu, a) = \frac{1}{1 + \frac{\pi \varepsilon_o \ell}{2\nu \, a \log(\ell/f_1)}} \tag{23}$$

The results are plotted in Figure 5, using the same parameter values as in Figure 3, with a 10-cm water gap. Similar to the results in Figure 3, a small fraction of the container area exposed by small holes, e.g., a = 1 mm or less, results in steady-state release rates that are almost as great as if the container were not present. Within the framework of the assumptions stated herein for the water-gap analysis, the maximum mass-transfer rate through a container with holes is no greater than the mass-transfer rate with no container. We expect that convective mixing in the water gap will steepen the curves in Figure 5 and bring the mass-transfer rate with holes even nearer the mass-transfer rate with no container.

CONCLUSIONS

The above calculations show that radionuclide transfer from multiple apertures can be significant and may approach or even exceed the mass transfer rate calculated for bare waste forms.

There are several possible implications for the repository program.

1. The U. S. Nuclear Regulatory Commission has not elaborated on the meaning of "substantially complete containment". This study indicates that radionuclide transfer rates can be significant from partially-failed waste containers, such as spent-fuel cladding. Diffusion through small apertures and cracks in the container can affect compliance with substantially complete containment.

2. Spent-fuel cladding may not be an effective barrier if many small perforations are anticipated.

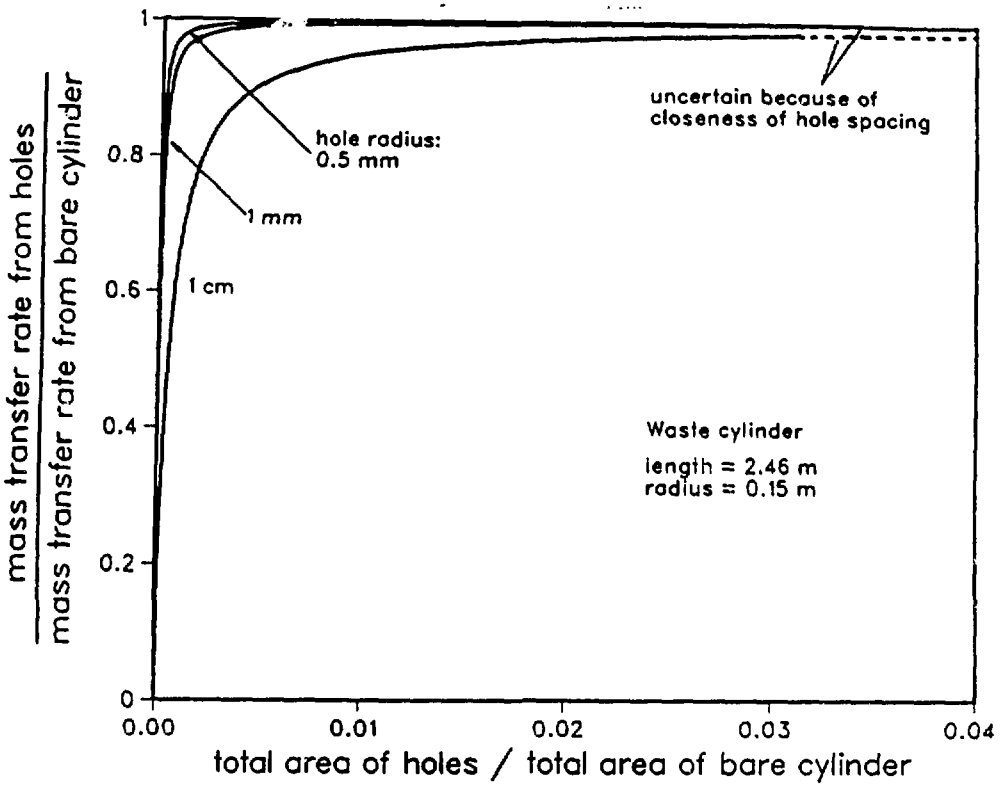


Figure 5. Radionuclide Transfer Rates from Multiple Holes Facing a Water Gap and from a Bare Waste Cylinder

3. Although partially-failed containers and container-corrosion products may present an important long-term barrier to release of radionuclides, small through holes have the potential of bypassing the effectiveness of these barriers.

4. To take credit for the long-term effectiveness of partially-failed containers as barriers to radionuclide release, details on the number and size of small penetrations and their growth with time will be needed.

5. For thin-walled containers with a sufficient number of small perforations, waste performance can be estimated by assuming waste form in direct contact with surrounding backfill or porous rock.

The analytical solutions presented here can be--and should be--verified by simple laboratory experiments.

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