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Nuclide Release from Penetrations in Radioactive Waste Container

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방사성 폐기물 저장용기 표면의 결함으로부터 핵종유출 연구

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Abstract

Nuclide release through penetrations in radioactive waste container is analyzed. Penetrations may result from corrosion or cracking and may be through the container material or through deposits of corrosion products. The analysis deals with the resultant nuclide release, but not with the way these penetrations occur. Numerical illustrations show that mass transport from multiple holes can be significant and may approach the mass transfer rate calculated from bare waste forms. Although partially-failed containers may present an important long-term barrier to release of radionuclides, numerous small holes on a container surface have the potential of bypassing the effectiveness of these barriers.

요 약

방사성폐기물 저장용기에 생긴 결함으로부터의 핵종유출을 분석하였다. 결함은 부식이나 균열으로부터 기인하며 저장용기의 재질이나 부식 부산물층을 관통하게 된다. 본 연구는 결함을 통한 핵종의 이동을 다루었으며 결함이 생기게 되는 방법에 관해서는 고려하지 않았다.
다수의 작은 결함으로부터의 핵종유출이 중요하며 저장용기가 없다고 가정한 폐기물고화체로부터의 핵종유출과 거의 비슷함을 보여준다. 비록 부분적으로 파손된 저장용기일지라도 핵종유출에 대하여서 장기간에 걸쳐 중요한 방벽의 역할을 하지만, 작은 결함의 수가 많아지면 방벽으로서의 효과가 줄어들게 된다.

1. Introduction

Radioactive waste will be disposed of in metal containers. A metal surface exposed to a corrosive environment may experience attack at a number of isolated sites. If the total area of these sites are much smaller than the surface area, the metal is experiencing localized corrosion. The rapidity with which localized corrosion can lead to the failure of a metal container and the extreme unpredictability of the time and place of attack, has led to a great deal of study of this phenomenon.

It is important to analyze the transport of species through penetrations in waste containers. The soluble radionuclides could dissolve inside the corroded container up to some limiting concentration and diffuse through the holes into the surrounding medium. In this paper the analytical solutions to predict rates of mass transfer through penetrations of specified size and geometry are discussed. Expressions for the diffusive mass transfer rates through apertures and numerical illustrations are presented.

2. Nuclide Transport Through a Hole

It is assumed that the nuclides are transported from the opening into the surrounding water-saturated medium by diffusion. The species in the waste form supplies a uniform concentration over an assumed time-invariant cross section of the aperture. The canister wall is treated as an infinite plane as shown in Figure 1.

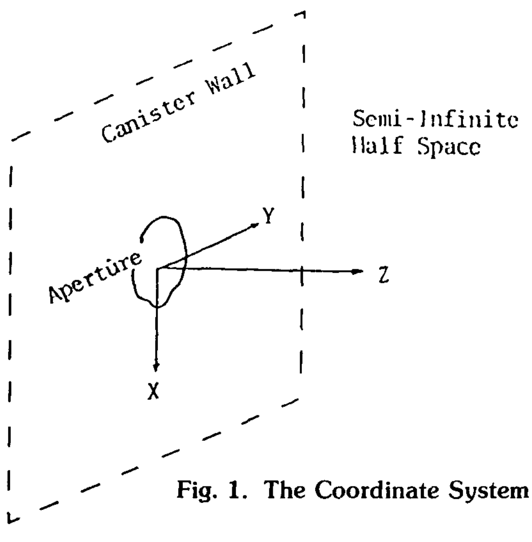

Fig. 1. The Coordinate System

The governing equations for the species concentration C (X,Y,Z) in the absence of radioactive decay at steady state are[1]

$$D \nabla^2 C = 0 \quad (1)$$

$$C = Cs, \text{ valid on aperture surface S} \quad (2)$$

$$-D\varepsilon \text{ grad } C = 0, \text{ valid on un-impaired canister surface} \quad (3)$$

D is the diffusion coefficient of the species. ε is the porosity of the water saturated medium. C and Cs are the species concentrations in the pore water and at the surface of the hole, respectively.

Eq.(2) furnishes the species concentration on the aperture surface S. It is assumed that the waste form volume is sufficiently large, so that one can ignore the loss of species through the opening. Eq.(3) indicates the absence of diffusive mass transport through the impermeable part of the canister surface. The time dependence is less important in the application to a repository setting where primarily the steady state solution is required. But the time dependent solution is important for the experimental validation of the theory.

The analysis is simplified by the introduction of the following dimensionless variables

$$X=\frac{X}{a_1},\ Y=\frac{Y}{a_1},\ Z=\frac{Z}{a_1},\ A=\frac{S}{a_1^2},\ C^*(X,Y,Z)=\frac{C(X,Y,Z)}{Cs} \quad (4)$$

Here a_1 is a characteristic dimension of the aperture and A is the dimensionless aperture surface. It is convenient to label the x,y coordinate separately from the z coordinate by writing

$$M=(X,Y),\ C^*=C^*(M,Z) \quad (5)$$

in the following. Eqs.(1) - (3) transform to

$$\nabla^2 C^*=0,\ |M|\langle\infty, Z\rangle 0 \quad (6)$$

$$C^*(M,0)=1,\ M\in A \quad (7)$$

$$\frac{\partial C^*(M,0)}{\partial Z}=0,\ M\notin A \quad (8)$$

At this point one must select a hole shape before a definitive solution is obtained. In the absence of specific information about failure configurations, a family of elliptically shaped apertures has been selected whose eccentricity can be parametrically adjusted to approximate the range from very long slits to a circular hole, see Figure 2.

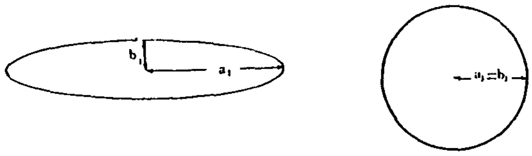

Fig. 2. Aperture Shapes

The solution to eqs.(6) - (8) in ellipsoidal coordinates is given by[1]

$$C^*(u) = F(u)/F(0), \quad 0<u<\infty \tag{9}$$

with

$$F(u) = \int_U^\infty \frac{du'}{\{(1+U')\ (\beta^2+U')U'\}^{1/2}} \tag{10}$$

where $\beta = b_1/a_1$ is the dimensionless semi-axis of the ellipse and the u coordinate describes the surface of the ellipsoid

$$\frac{X^2}{1+U} + \frac{y^2}{\beta^2+U} + \frac{z^2}{U} = 1 \tag{11}$$

For an irregularly shaped aperture, it is intuitively apparent that the total mass flux is bounded by the flux through inscribed and circumscribed elliptical apertures, see Figure 3.

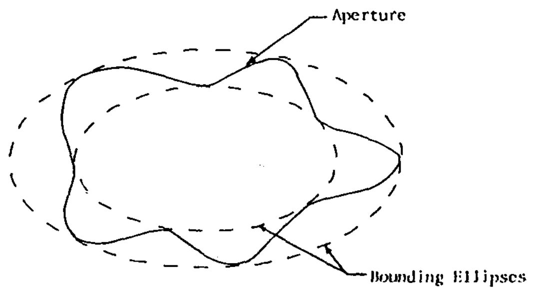

Fig. 3. Bounding Contours

The isopleths, in ellipsoidal coordinates, can be computed from the solution (9) in terms of an elliptic function. In the limiting case of a circular opening, for which $\beta = 1$, the solution takes the elementary form

$$C^*(u) = \frac{2}{\pi} \tan^{-1}\left(\frac{1}{\sqrt{u}}\right) \tag{12}$$

or transformed to cartesian coordinates with $r^2 = x^2 + y^2$

$$C^*(r,z) = \frac{2}{\pi} \sin^{-1}\left(\frac{2}{\sqrt{z^2+(1+r)^2}+\sqrt{z^2+(1-r)^2}}\right) \tag{13}$$

The steady-state isopleth surfaces are shown in the domain r>0, z>0 in Figure 4. At r = 0 and z = 0.5, the steady-state concentration is less than 70 percent of 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 4 are the diffusion paths, parallel to the concentration gradients. Near the edge of the hole the diffusive mass transfer is most intense.

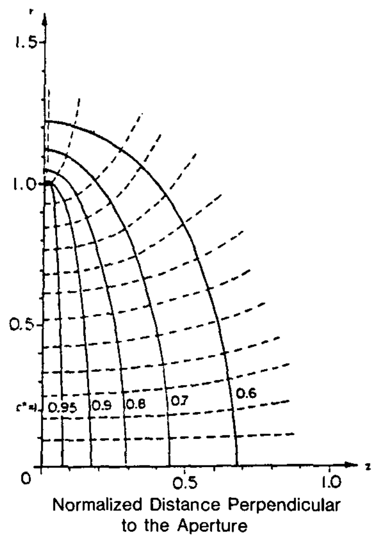

Fig. 4. Surfaces of Non-dimensional Concentrations and Diffusional Paths Near an Aperture

The steady-state dimensional mass release rate through a circular hole of radius a_1 is

$$\dot{m}=a_1^2\int_0^1 2\pi r\left[-D\varepsilon\frac{Cs}{a_1}\frac{\partial C^*(r,z)}{\partial z}\bigg|_{z=0}\right]dr=4D\varepsilon a_1 Cs \quad (14)$$

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.

3. Multiple Holes in a Cylindrical Waste Container

If multiple holes are present on a cylindrical waste container, the total mass transfer rate is the sum of contributions from all holes. (Fig. 5)

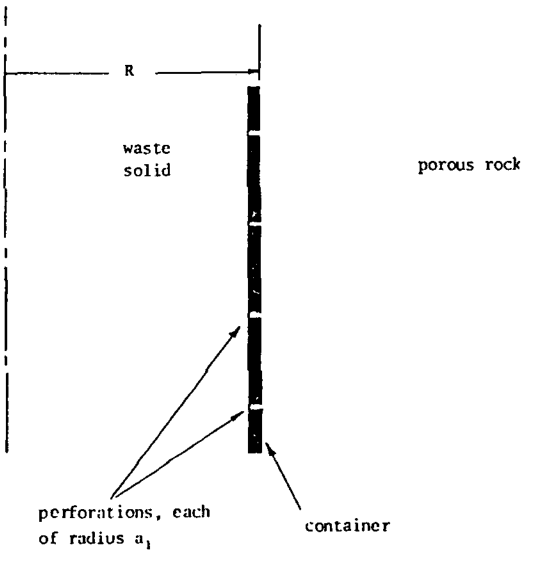

Fig. 5. Multiple Holes

For this sum to be obtained from the single-hole equation, 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 apertures. From eq.(13), it is seen that at $z=0$, $r=10$, then $C^*=0.064$, and the concentration has fallen to negligible level about 10 hole radii away.

Consider a square-grid distribution on the flat rectangular surface that is obtained by unfolding a cylindrical container. If the radius and the length of the cylinder is R and L, the number of holes that can fit in the rectangular surface is

$$N=2\pi RL/P^2 a_1^2 \quad (15)$$

where p is the pitch of the distribution of holes normalized to the hole radius. The contribution from one hole to another is bounded by the following approximation:

$$C^*(r,0)=2/(\pi r), \quad r\geqq 10 \quad (16)$$

Then, the contribution from N holes to the center hole is approximately

$$\Delta C^*\leqq\frac{4}{\pi p}(1+1/\sqrt{2})\sqrt{2\pi RL/p^2a_2^2} \quad (17)$$

If one assumes $\Delta C^*=0.064$ as acceptable for superposition to apply, the allowable pitch for 1-mm radius holes on a cylindrical container with a radius of 0.15 m and a length of 2.46 m is 0.22 m. Thus one can place 45 holes of 1-mm radius.

4. Mass Transfer Rate from Multiple Independent Holes

To get the acceptable working definition of independent holes, the minimum pitch needed to consider the holes as independent is used. If N holes are independent of each other, linear superposition results in a total mass transfer rate $\dot{m}_a$ through these apertures as follows:

$$\dot{m}_a=4ND\varepsilon a_1C_s \quad (18)$$

If the waste container is not present, the steady-state mass transfer rate $\dot{m}_c$ from a waste form cylinder of radius R and length L in contact with the medium of porosity ε is[2]

$$m_c = 2\pi D\varepsilon C_s\ L/\log(L/R) \quad (19)$$

From eqs. (18) and (19), the ratio $\alpha(N, a_1)$ of the mass-transfer rates with and without a perforated container is

$$\alpha(N, a_1) = \frac{2Na_1\ \log(L/R)}{\pi L} \quad (20)$$

The results are plotted in Figure 6. Chambré et al.[1] applied linear superposition to show that a waste package with a large number of small holes could have a comparable mass release rate with a bare waste form of the same overall size. For 1-mm radius holes, however, roughly 50 independent holes can be superimposed with eq. (20) as discussed in the previous section. Since the total area of the 50 holes of 1-mm radius is 1.57 cm^2, it corresponds to a fraction of 7×10^{-5} of the container surface. An assumption often made in codes for radionuclide release calculation, for example the WAPPA code[3], is that the diffusional transport through holes of known area in the container is given by the diffusional transport from a bare solid waste multiplied by the ratio of hole area to the total container surface area. Also plotted in Figure 6 is the analogous calculation based on the corroded-area proportionality approximation assumed by the WAPPA code. However the steady state release of radionuclides from multiple interacting holes cannot be estimated by eq. (20).

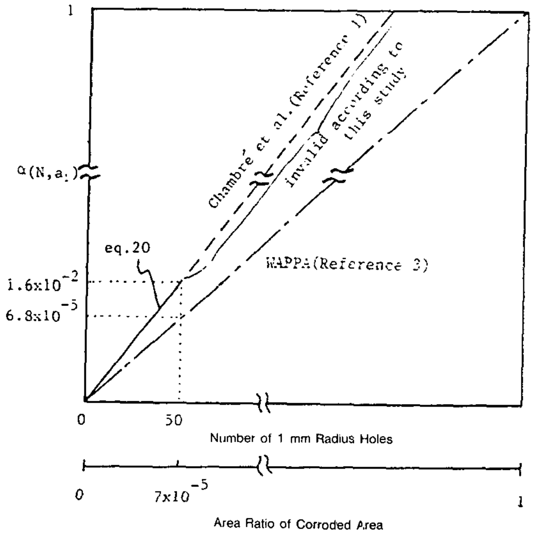

Fig. 6. Nuclide Transfer Rates from Multiple Holes of 1 mm Radius on a Cylindrical Container with a Radius of 0.15 m and a Length of 2.46 m

5. Conclusions

Diffusion through small apertures and cracks in the container may affect container performance. Numerical illustrations show that radionuclide transfer from multiple apertures can be significant. Linear superposition can be applied up to 50 independent holes of 1 mm radius. The corroded-area proportionality approximation will generate non-conservative results for multiple independent holes.

However partially-failed containers and container-corrosion products may present an important long-term barrier to release of radionuclides. 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 holes and their growth with time will be needed.

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