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MASS TRANSFER FROM PENETRATIONS IN WASTE CONTAINERS*

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BNL 40645

BNL--40645

DE88 005604

Paper presented at "Scientific Basis for Nuclear Waste Management-XI" November 30-December 3, 1987, Boston, Massachusetts (Proceedings will be published as Vol. 112 of the Materials Research Society Symposium Proceedings Series).

ABSTRACT

Recent studies have indicated that localized corrosion of a relatively small area of a waste container may impair the containment function to such an extent that larger releases may be possible than from the bare waste form. This would take place when a large number of holes coexist on the container while their concentration fields do not interact significantly with each other. After performing a steady state analysis of the release from a hole, it is shown that much fewer independent holes can coexist on a container surface than previously estimated. The calculated radionuclide release from multiple independent holes must be changed accordingly. Previous analyses did not proceed to a correct application of the linear superposition principle. This resulted in unacceptable physical conclusions and undue strain on the performance assessment necessary for a container licensing procedure. The paper also analyzes the steady state release from penetrations of finite length and whose concentration fields interact with one another. The predicted release from these penetrations is lower than the previously calculated release from holes of zero thickness. It is concluded here that the steady-state release from multiple holes on a waste container can not exceed the release from the bare waste form and that multiple perforations need not be a serious liability to container performance.

INTRODUCTION

One of the most likely mechanisms by which nuclear waste containers may lose their integrity is through localized corrosion of the container walls. In a water-saturated repository this may result in diffusion of radionuclides from the waste into the surrounding medium.

Reports are available regarding the extent to which penetrations of small radii (with respect to container dimensions) may affect container performance. The most recent studies, by Chambrè and co-workers, indicate that localized corrosion of a relatively small area of the container may impair the containment function to such an extent that larger releases may be possible than from the unprotected waste form [1,2] (Figure 1). The reason given is that the steady-state diffusion flux from a small hole with a uniform radionuclide concentration is extremely large at the edge of the hole. Thus for a given area of attack, the larger the number of holes the smaller is their radius and the larger is the total predicted release rate. Virtually the same analysis had been carried out by Rae [3]. Although Rae's conclusions were that "...even a canister with appreciable perforations will release only a very small fraction of its contents per year and can still be regarded as a reasonable container," his numerical estimates indicate the contrary. In Rae's calculations a localized attack equivalent to 3.14 cm^2 on a surface of $7x10^4$ cm^2 or larger results in a

*This work was performed under the auspices of the U. S. Department of Energy.

fractional release rate of 10^{-6} per year if the attack is concentrated on one hole of 1-cm radius and in a fractional release of 10^{-3} per year if the release is from one-thousand holes of 1-mm radius.

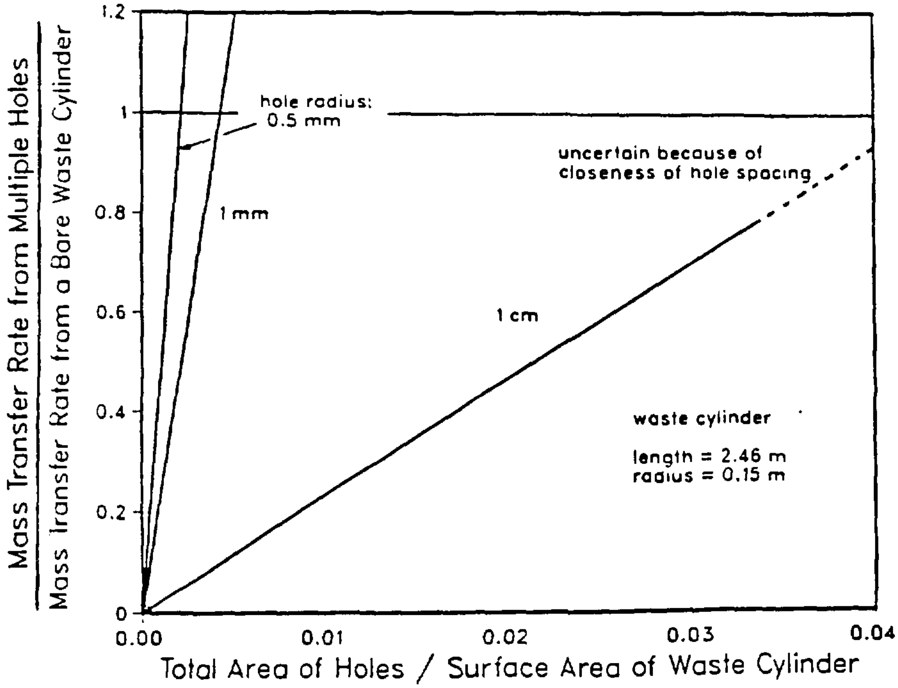

Figure 1. Radionuclide Transfer Rate From Multiple Non-interacting Holes and a Bare Waste Cylinder (Adapted from [1])

The view held in the literature that small amounts of perforated area may sensibly affect container performance is troublesome from a physical as well as from a licensing point of view. One cannot readily accept that if the attack is properly localized, more material would be lost than from the unclad waste form (Figure 1). In a heat transfer analogy, this would indicate that by covering a heat source of constant temperature with an insulating material and drilling then an appropriate number of holes in this sheathing one might extract more heat than from the bare source. On the other hand, if these analyses were acceptable, small holes would have the potential of bypassing the effectiveness of container corrosion resistance. For licensing purposes this would complicate the analysis of container performance in a profound manner as details on the number, distribution, and size of small penetrations would be needed along with their growth in time. Details of the Chambrê analyses have already been implemented in source term codes for waste package performance [4].

In this paper we propose to show that the steady-state release of radionuclides from multiple holes cannot exceed the release from a bare waste form and that multiple perforations need not be a serious liability to container performance.

Analytical results

Both Chambrê's and Rae's results rest on a steady-state mass transfer analysis from a hole on a flat surface into a surrounding medium. The surface is taken to be of infinite extent and the transport of mass is modeled to occur by diffusion only. With reference to the coordinate system of Figure 2, if $C(r,z)$ is the concentration of a given chemical species outside the hole, and C_s is the saturation concentration of that species at the surface of the hole, the distribution in space is determined by solving the following equation and boundary conditions:

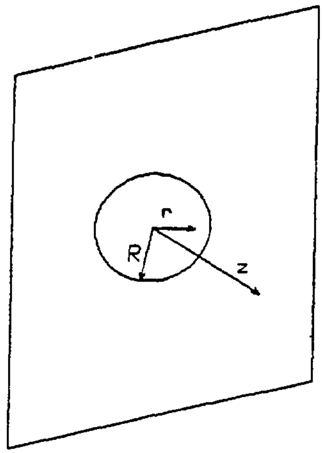

Figure 2. The Coordinate System

$$\nabla^2 C(r,z) = 0 \qquad r,z \geq 0 \tag{1}$$

$$C(r,0) = C_s \qquad r < R \tag{2}$$

$$\partial C(r,z)/\partial z = 0 \qquad r > R;\ z = 0 \tag{3}$$

The problem has mathematical analogs in the theory of electrostatics and in fluid dynamics. Its solution can be found in standard classical texts [5, 6]. In particular, if C^* is the concentration normalized to C_s, it is:

$$C^*(r,z) = \frac{2}{\pi}\ \sin^{-1} \frac{2\ R}{[z^2 + (R+r)^2]^{1/2} + \left|[z^2 + (r-R)^2]^{1/2}\right|} \tag{4}$$

from which:

$$\left.\frac{\partial C^*}{\partial z}\right|_{z=0} = -\ \frac{2}{\pi}\ \frac{1}{(R^2 - r^2)^{1/2}} \qquad r < R \tag{5}$$

Introducing D, an effective diffusion coefficient, the following interesting expression for the integrated flux of material, q, out of a single hole is obtained:

$$q = 4 D C_s R \tag{6}$$

i.e., the integrated flux is proportional to the radius and not to the area of the hole. A fixed area, A, of localized attack on the container can accommodate a number, N, of holes of the same radius which is given by:

$$N = A / p^2 R^2 \tag{7}$$

where p is the pitch of the distribution of holes normalized to the hole radius. Thus, if these holes are independent from each other, linear superposition of contributions results in a total integrated flux, Q, given by:

$$Q = 4 D C_S \frac{A}{p^2 R} \tag{8}$$

i.e, for a given area, the smaller the radius of the independent holes (and therefore the larger their number) the larger is the total release, if the normalized pitch is held constant. This accords indeed with the findings of Chambrè and Rae; the crux of the problem, however, is an acceptable working definition of "independent holes," so that the superposition principle may be utilized.

"Independent" Perforations

The smallest spacing between holes for modeling any two holes as behaving independently from each other was taken to be equal to 10-hole radii in previous studies. It is in fact $C^*(10R,0)=0.064$. By taking a pitch of 10-hole radii for the distribution of holes on a container surface, hundreds or thousands of "independent" holes can be accommodated.

In contrast with previous studies, we find that only a few tens of holes may coexist on a container before interfering with each other. In particular, we find that the minimum pitch needed to consider the holes as independent can not be based on the interaction between two holes, as was previously done, but must take into account the total number of holes present on the surface.

It should be noted that the contribution from one hole to another is bounded by the following inequality:

$$C^*(r,0) \geq \frac{2}{\pi} \frac{R}{r} \qquad r \geq 10 R \tag{9}$$

From Equation (9) $C^*(10R,0) \approx 0.064$. If N+1 circular holes of the same radius are distributed along a straight line the contribution, $\Delta_N C^*$, by N holes to the end hole is:

$$\Delta_N C^* = \frac{2}{\pi\, p_N} \sum_{n=1}^{N} \frac{1}{n} \qquad r \geq 10\ R \tag{10}$$

where "p_N" is the pitch of the distribution normalized to the hole radius, R. Thus, if holes are added to the distribution, one has to increase the pitch as a harmonic series in order to keep $\Delta_N C^*$ constant. If one assumes $\Delta_N C^*=0.064$ as acceptable for superposition to apply, as it is suggested in [1], the allowable pitch varies as follows with the number of holes: $p_N=10$ for N=1, $p_N=29$ for N=10, $p_N=52$ for N=100, and $p_N=74$ for N=1000: a logarithmic growth. The total length, L, needed to accommodate (N+1) non-interacting holes grows faster with N since $L=p_N RN$.

The rate of increase of the allowable pitch with the number of holes is stronger still for a square-grid distribution of holes. Consider one such distribution on the flat rectangular surface that is obtained by unfolding a cylindrical container. Focusing our attention on the maximum inscribable circle on this surface, the number of holes that can fit in this circle of radius L is roughly $N=\pi L^2/R^2 p_N^2$. The contribution from these holes to the center hole is approximately:

$$\Delta_N C^* = \frac{4\,(N/\pi)^{1/2}}{p_N} \tag{11}$$

If one again assumes $\Delta_N C^*=0.064$ as acceptable for superposition to apply, the allowable pitch varies as follows with the number of holes:

$$p_N \geq 35\ N^{1/2} \tag{12}$$

Accordingly, the area, A, needed to accommodate N non-interacting holes increases even faster with N. It can be shown that $A \geq 35^2 N^2 R^2$. As an example, in order to accommodate 1,000 non-interacting holes each of 1-mm radius, one needs a surface area of the order of 1,225 m^2. This is an extremely large area compared to the surface area of a waste container.

If one considers a waste container of height H=2.46 m and radius $R_{WF}=0.15$ m as in Figure 1, the lateral area of the container is 2.31 m^2. The area of the maximum inscribable circle on this surface is 0.70 m^2. On the latter area one may fit no more than N=0.025/R independent holes (R measured in meters). Thus, if R=0.001 m, one can at most place 25 holes within the circle. Analogously, one may not fit more than 65 holes within the circle that circumcribes the whole surface. Therefore roughly 50 holes only of 1-mm radius can fit on the given surface without significantly interacting with each other, whereas the analysis of Chambrè allows 23,000 independent holes. Since the total area of the 50 holes is 1.6×10^{-4} m^2, it corresponds to a fraction of 7×10^{-5} of the container surface. Applying the following equation adapted from the work of Chambrè:

$$F_N = \frac{2\,N\,R}{\pi\,H} \log(H/R_{WF}) \tag{13}$$

the release rate, F_N, from the 50 holes normalized to the bare waste form case is estimated to be F_{50}=0.015, slightly more than 1%.

One realizes therefore that much fewer independent holes may fit on a given surface than previous analyses had estimated and that the validity of Figure 1 is restricted to a very small region of its lower left-hand corner. So small, in fact, that Figure 1 does not provide enough definition.

Numerical Modeling of Finite-Length Interacting Holes

General Case

When holes are distributed sufficiently close to one another, they can no longer be modeled as behaving independently. On the other hand, with reference to a 2-D distribution of holes on a flat surface, it can be observed that symmetry planes can be drawn extending from around each hole into the porous medium. These planes define channels with similar concentration fields. Thus it is advantageous to confine the analysis to one such combination of hole+channel. We have performed the analysis by considering the channel to be of cylindrical cross section and of radius R equaling the pitch of the distribution. In order to obtain a generic expression for the total release rate the following scheme was devised:

With reference to Figure 3, assume a wall of thickness t with a cylindrical hole (having its axis perpendicular to the wall) of radius r. Let two planes parallel to the wall at distances L from the wall be kept at concentrations C_1 and C_2. Let the space, including the pit, be filled with a diffusive medium in which the diffusant has a diffusion coefficient D. Define a cylindrical unit cell of radius R coaxial with the pit.

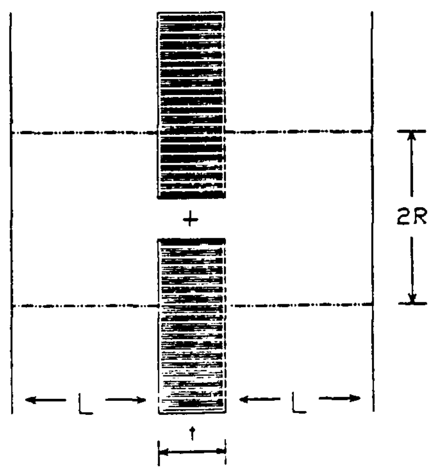

Figure 3. Example of Cell for Transport Calculations

If the wall has N pits per unit area, then R is defined by:

$$N \pi R^2 = 1 \tag{14}$$

At steady-state, the concentration distribution is symmetrical about the mid-plane of the wall. The latter is an iso-concentration surface at:

$$C_0 = 0.5\ (C_1 + C_2) \tag{15}$$

Therefore we can concentrate on one-half of the space, between the mid-plane of the wall and the surface at C_2. The result of interest is the steady-state mass flow between those planes.

Let $\ell = t/2$, and let L be the distance between the wall and the plane at C_2. In a form reminiscent of the treatment of contractions [7] in hydraulics, we can define an entrance region term as follows:

$$q = \frac{D\ (C_0 - C_2)}{\dfrac{\ell}{\pi r^2} + \dfrac{L}{\pi R^2} + f(\ell, L, r, R)} \tag{16}$$

where q is the mass transport rate for the unit cell.

The correction term f can be re-defined to be of the form:

$$f(\ell, L, r, R) = \frac{k(\ell/r, L/r, R/r)}{\pi r} \tag{17}$$

In order to obtain values for the parameter k and understand its behavior one can solve the equivalent heat conduction problem with adiabatic boundary conditions at the intact wall surface and between contiguous cells and with fixed concentrations C_0 and C_2 at the remaining boundaries. We modeled this unit cell by using the heat transfer code HEATING5 [8] in cylindrical geometry. The results of several runs with different combinations of the parameters ℓ/r, L/r, and R/r are listed in Table 1.

The correction factor k goes to zero when R tends to r, as is to be expected, and its values are always positive. This means that a release rate expression of the form:

$$q = \frac{D\ (C_0 - C_2)}{\dfrac{\ell}{\pi r^2} + \dfrac{L}{\pi R^2}} \tag{18}$$

will always overpredict the radionuclide flow. In particular, if the aspect ratio of the hole, ℓ/r, is equal or larger than, say, 0.5, the overprediction of the flux afforded by Equation (18) is not significant as the contribution from the k-term (Table I) is roughly equal to or smaller than the ℓ-term in Equation (17). This is the case to be expected in the applications.

Table I. Values of the Parameter k in Equation (17)

ℓ/r	L/r	R/r	k
3	5	20	0.524
3	10	20	0.512
3	20	20	0.529
3	30	20	0.516
3	5	5	0.443
3	10	5	0.439
3	20	5	0.43
3	30	5	0.425
3	5	3	0.347
3	10	3	0.345
3	20	3	0.343
3	30	3	0.342
3	5	2	0.217
3	10	2	0.217
3	20	2	0.216
3	30	2	0.216
3	5	1.5	0.116
3	10	1.5	0.116
3	20	1.5	0.116
0.5	2	1.5	0.146
0.5	7	1.5	0.139
0.5	17	1.5	0.124
0.5	2	2	0.279
0.5	7	2	0.257
0.5	17	2	0.237
0.5	2	5	0.525
0.5	7	5	0.415
0.5	17	5	0.293
0.5	2	10	0.566
0.5	7	10	0.528
0.5	17	10	0.48

For the practical task of solving for the concentration field away from an interacting hole, one can use a numerical scheme implementing Equation (19) as a boundary condition at the first grid line outside the container surface. Once the distance of this line, L, from the container surface is fixed, C_2 and the concentration values farther away can be obtained by applying an iterative solution strategy to the discretized set of equations representing the original diffusion equation. This approach avoids the numerically exacting task of calculating the concentration field inside and near the surface of the hole, which would have to be performed for every value of the hole radius and depth.

Finite-Length Non-Interacting Holes

A further result that can be obtained starting from Equations (16) and (17) and from the results of Table I is a bounding expression for the flux from independent holes of a finite length, ℓ. By letting L/R go to zero in Equation (16) and setting further C_2=0 at the container surface, it can be seen that the flux is bounded by the expression:

$$q = \frac{C_0 \, D \, \pi \, r^2}{\ell + 0.5 \, r} \tag{19}$$

A comparison of Equation (19) with Equation (4), indicates that for finite length holes the release is proportional to the area of the hole and not to its radius. Furthermore Equation (19) predicts a lower release rate than Equation (4) as long as the aspect ratio of the hole, ℓ/r, is greater than 0.3, a circumstance that is to be expected in practical situations.

CONCLUSIONS

Recent studies have indicated that localized corrosion of a relatively small area of a waste container may impair the containment function to such an extent that larger releases may be possible than from the bare waste form. This would take place when a large number of holes coexist on the container without interacting with each other. After performing a steady-state analysis of the release from a hole, it is shown that much fewer independent holes can coexist on a container surface than previously estimated. The calculated radionuclide release from multiple independent holes must be changed accordingly. Previous analyses did not proceed to a correct application of the linear superposition principle. This resulted in unacceptable physical conclusions and undue strain on the performance assessment necessary for a container licensing procedure. We have analyzed the steady state release from holes of finite length and whose concentration fields interact with one another. The predicted release from these holes is lower than the predicted release from "mathematical" holes of zero thickness, as they were modeled in previous studies. It is concluded here that the steady-state release from multiple holes on a waste container can not exceed the release from the bare waste form and that multiple perforations need not be a serious liability to container performance.

ACKNOWLEDGEMENT

During the preparation of this paper we benefited from useful discussions with Dr. T. Sullivan, a member of our group at BNL.

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