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Radionuclide Mass Transfer Rates from a Pinhole in a Waste Container for an Inventory-Limited and a Constant Concentration Source

Taux de transfert de masse, par une piqûre dans un conteneur de déchets, de radionucléides issus d'une source présente en quantité limitée et à concentration constante

D.M. LeNeveu

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D.M. LeNeveu

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ABSTRACT

Analytical solutions for transient and steady state diffusive mass transfer rates from a pinhole in a waste container are developed for constant concentration and inventory-limited source conditions. Mass transport in three media are considered, inside the pinhole (medium 2), outside the container (medium 3) and inside the container (medium 1). Simple equations are developed for radionuclide mass transfer rates from a pinhole. It is shown that the medium with the largest mass transfer resistance need only be considered to provide a conservative estimate of mass transfer rates.

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TAUX DE TRANSFERT DE MASSE, PAR UNE PIQÛRE DANS UN CONTENEUR DE DÉCHETS, DE RADIONUCLÉIDES ISSUS D'UNE SOURCE PRÉSENTE EN QUANTITÉ LIMITÉE ET À CONCENTRATION CONSTANTE

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RÉSUMÉ

L'auteur élabore dans la présente communication des calculs analytiques du taux de transfert de masse par diffusion, en régime stationnaire et transitoire, par une piqûre dans un conteneur de déchets, à partir d'une source en quantité limitée et de concentration constante. Le transport de masse est pris en compte dans trois milieux : à l'intérieur de la piqûre (milieu 2), à l'extérieur du conteneur (milieu 3) et à l'intérieur du conteneur (milieu 1). La représentation du taux de transfert de masse par une piqûre est établie par des équations simples. On démontre qu'il suffit de tenir compte du milieu offrant la plus grande résistance au transfert de masse pour produire des estimations prudentes des taux de transfert de masse.

EACL
Laboratoires de Whiteshell
Pinawa (Manitoba)
Canada R0E 1L0
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CONTENTS

1. INTRODUCTION

Concepts for the geological disposal of radioactive waste usually include isolation of the waste inside a durable container. Fabrication defects could cause a small number of these containers to have pinholes through which radionuclides could escape. Several studies have been done on the mass transfer of radionuclides through holes in waste containers. Rae (1985) developed equations for steady state mass transfer rate for a two medium case, one inside the container and the other, outside. Chambré et al. (1986) developed analytical equations for steady state mass transport for a single medium, external to the container, for differing hole geometries and developed asymptotic approximations to transient transport. Radioactive decay was treated explicitly only by Chambré et al. (1986). DePaoli and Scott (1993) developed a numerical model for the two medium case for both transient and steady state conditions. They emphasized the importance of the mass transport properties internal to the container in determining the overall mass transfer rate from a pinhole. Pescatore and Sastre (1988) considered the effects of multiple holes under steady state conditions. They examined mass transport inside the pinhole, inside the container and outside the container for the same medium throughout.

This study differs from the above-mentioned studies in that mass transport of radionuclides is developed considering the properties of the interior of the pinhole (medium 2), the exterior of the container (medium 3) and the interior of the container (medium 1), for both transient and steady state conditions, and for two types of source boundary conditions, inventory-limited and constant concentration. The inventory-limited boundary condition applies to readily leachable radionuclides that would be released to the interior of the container soon after ingress of water. The constant concentration boundary condition applies to radionuclides whose concentrations in the water inside the container are determined by their solubility.

In this study, expressions for both transient and steady state mass transfer rates for inventory-limited and constant concentration source boundary conditions are developed. An inventory-limited boundary condition refers to an initial deposition of a radionuclide in the medium that becomes depleted by mass transport from the medium. First, mass transport through the pinhole is considered assuming that the mass transfer resistance inside and outside the container are negligible in comparison. This means that the concentration will be spatially uniform inside the container and that concentration appearing at the exit from the pinhole will be quickly transported away. Secondly, mass transport outside the pinhole is considered assuming the mass transfer resistance inside the container and the pinhole are negligible in comparison. This means that the concentration will be spatially uniform inside the container and inside the pinhole. Although mass transport inside and outside the pinhole are considered separately, the equations for the medium with the largest mass transfer resistance can be used for the overall mass transfer rate from the pinhole. This approach is accurate when the mass transfer resistance of one medium is much larger than the mass transfer resistances of the other media and gives an overestimate of maximum mass transfer rates otherwise. In waste management studies, where compliance is measured quantitatively with respect to regulatory criteria, using overestimates of mass transfer rates is called a conservative approximation (Goodwin et al. 1994) and is normally considered to be acceptable provided that the overestimate is not unreasonably large. Finally, using the concept of addition of mass transfer resistances, an expression is obtained for pseudo-steady state conditions that applies to all the media in the system simultaneously.

Radioactive decay of a single radionuclide is accounted for, but not for a decay chain. If mass transport for a decay chain is of interest, it could be assumed that the influence of a precursor on mass transfer release rates are small. This is a reasonable assumption because the residency times of a radionuclide in the very small distances inside the pinhole or in the region of influence around the pinhole are likely to be so small that ingrowth would not have a large effect on concentration gradients except for radionuclides with very short half-lives. Such radionuclides are normally considered to be in secular equilibrium with a parent and their mass transfer rates can be determined directly from the parent (Goodwin et al. 1994).

2. MASS TRANSPORT INSIDE THE PINHOLE (MEDIUM 2)

2.1 SYSTEM DESCRIPTION

The assumed geometry of the pinhole is illustrated in Figure 1.

Mass transfer rates from a tortuous pinhole with a smaller cross-section at some points along the length are likely to be smaller than from a pinhole with the simplified geometry of Figure 1. Thus it should be conservative to simplify the geometry as shown in Figure 1 provided the largest cross sectional area along the length of the pinhole is used.

It is assumed that the entire system shown in Figure 1 is water saturated. The mass transport media in the system would depend on the particular waste container emplacement design. Medium 1, inside the container, could be a porous infilling material such as glass beads or sand, or the container could be unfilled. Medium 2, inside the pinhole, could be unfilled or be filled with porous corrosion product or extruded backfilling material. Medium 3, outside the container, would be a porous backfilling material such as a mixture of clay and sand.

If the concentration inside the pinhole is considered to be uniform in directions perpendicular to the axis of the hole, then the one-dimensional mass balance equation can be used for the simplified geometry of Figure 1:

$$K_2 \frac{\partial C}{\partial t} = D_2 \frac{\partial^2 C}{\partial x^2} - \lambda \ K_2 C. \tag{1}$$

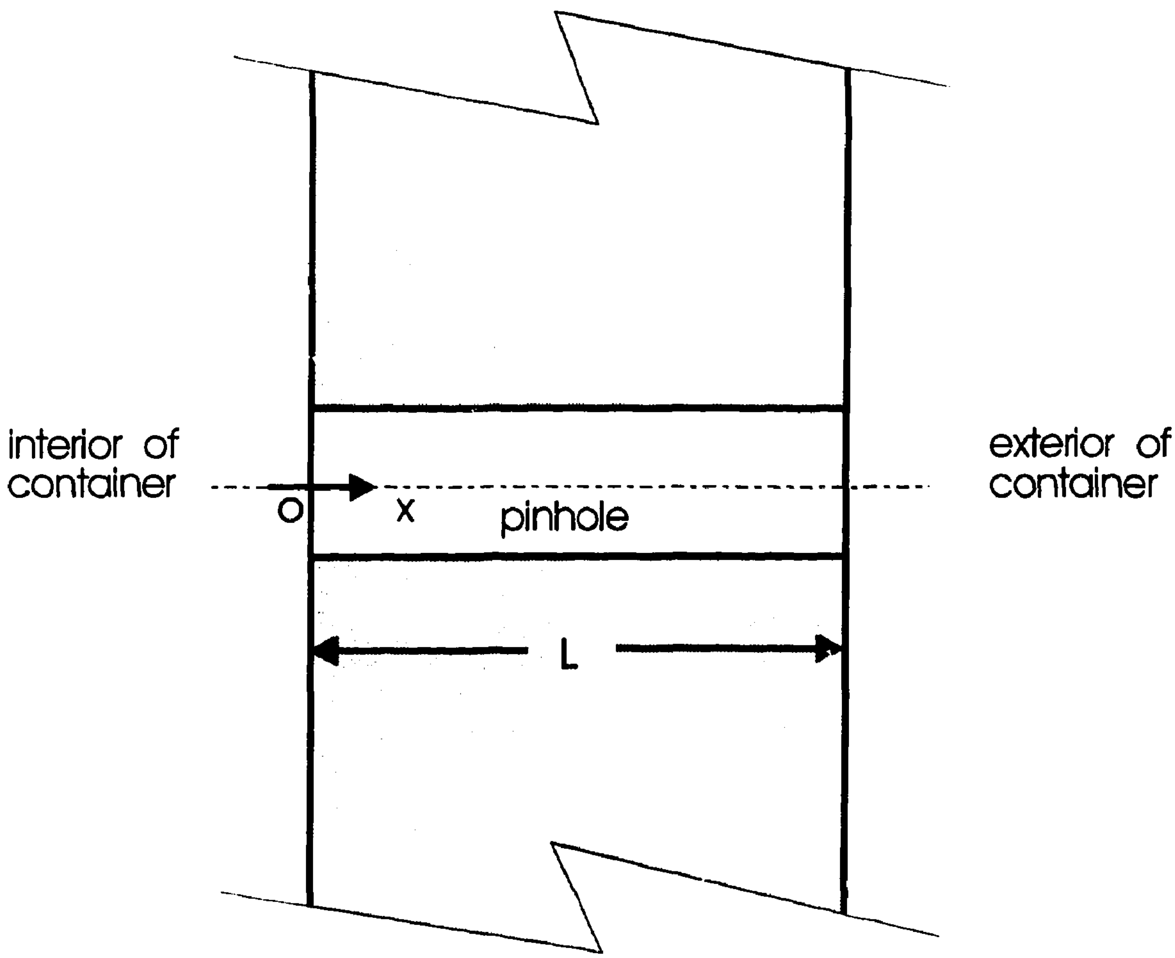

FIGURE 1: Side View of the Pinhole.

Here, $C(x,t)$ is the concentration of radionuclides in the pore water in the pinhole, D_2 is the total intrinsic diffusion coefficient (Cook 1988) in the pinhole, K_2 is the radionuclide capacity factor (Oscarson 1994) in the pinhole, λ is the radionuclide decay constant, x is the distance along the axis of the pinhole, t is time and the subscript, 2, indicates medium 2. In terms of more fundamental parameters, the total intrinsic diffusion coefficient, D_i, in the absence of surface diffusion, can be expressed as:

$$D_i = D_{pi}\eta_{ei} \tag{2}$$

where D_{pi} is the pore diffusion coefficient, η_{ei} is the porosity available for diffusion and i is the index for the medium, with i=1 referring to the medium inside the container, i=2 referring to the medium inside the pinhole and, i=3 referring to the medium outside the container. The capacity factor, K_i, can be expressed as:

$$K_i = \eta_{ei} + \rho_i K_{di} \tag{3}$$

where ρ_i is the bulk dry density of the medium and K_{di} is the radionuclide distribution coefficient (Oscarson 1994, Cook 1988).

Inside the pinhole and elsewhere, it is assumed that the fluid motion is negligible so that convective transport can be ignored.

2.2 MASS TRANSPORT THROUGH THE PINHOLE FOR AN INVENTORY-LIMITED SOURCE

An inventory-limited source refers to a finite amount of radionuclide, I, uniformly distributed inside the container at t=0. The radionuclide is distributed between two phases with part in aqueous solution and the remainder sorbed onto the solid surfaces inside the container. Assuming the mass transfer resistance inside the pinhole is much larger than inside the container the radionuclide remains uniformly distributed such that there is no spatial variation of concentration inside the container.

For an inventory-limited source for mass transport inside the pinhole, the boundary condition at x=0 is:

$$K_1 V \frac{\partial C}{\partial t} = D_2 \frac{\partial C}{\partial x} A - \lambda K_1 V C. \tag{4}$$

Here, V is the internal volume of the container, A is the cross-sectional area of the pinhole and K_1 is the capacity factor in medium 1. In the absence of sorption, the void volume inside the container is given by, $K_1 V$. Note that, even though mass transport is considered in medium 2 only, parameters pertaining to the interior of the container (medium 1) appear through the application of the boundary condition.

Assuming the mass transfer resistance inside the pinhole is much larger than outside the container, mass appearing at the exit of the hole will quickly diffuse away. Accordingly, the boundary condition at $x=L$, where L is the length of the pinhole, is:

$$C(L,t)=0. \tag{5}$$

The initial condition at t=0 in the pinhole is:

$$C(0,0)=\frac{I}{K_1V}, \quad C(x,0)=0, \quad x>0. \tag{6}$$

The radionuclide mass transfer rate from the pinhole, $F_2(L,t)$, can be determined from:

$$F_2(L,t)=-D_2A\frac{\partial C}{\partial x}\Big|_{x=L}. \tag{7}$$

The solution for $F_2(L,t)$ can be determined by standard methods (Carslaw and Jaeger 1959),

$$F_2(L,t)=\frac{2D_2AI}{L}\sum_{k=1}^{\infty}\frac{\beta_k\exp\left(\frac{-\beta_k^2D_2t}{L^2K_2}-\lambda t\right)}{(LAK_2+K_1V)\sin(\beta_k)+K_1V\beta_k\cos(\beta_k)} \tag{8}$$

where β_k are all the positive roots, in increasing order, of:

$$\beta\tan(\beta)=\frac{K_2LA}{VK_1}. \tag{9}$$

If $K_2LA/(VK_1) << 1$ then β_k can be approximated by:

$$\beta_k=\delta_{k1}\sqrt{\frac{K_2LA}{K_1V}}+(k-1)\pi, \qquad k\geq1 \tag{10}$$

Here, δ_{kj} is the Kronecker delta. For $t > L^2K_2/D_2$ and $K_2AL/(K_1V) << 1$, $F_2(L,t)$ reduces to:

$$F_2(L,t)=\frac{D_2AI}{K_1VL}\exp\left(-\frac{D_2At}{K_1VL}-\lambda t\right). \tag{11}$$

For small times, $t \leq L^2K_2/D_2$, the mass transfer rate as predicted by Equation (8) will be less than that given by the evaluation of Equation (11) during the duration of the transient ($0 < t \leq L^2K_2/D_2$) because the radionuclide takes some time to diffuse down the length of the pinhole. For most waste management studies the duration of the transient is too short to be of any consequence; thus the use of Equation (11) throughout the entire time domain should be adequate. Should it be of interest to evaluate the transient behaviour more exactly, Equation (8) could, in principle, be used. However, Equation (8) converges very slowly during the transient time domain. It would be more practical to numerically invert (Talbot 1979) the Laplace transform solution given by:

$$F_2(L,s) = \frac{2D_2q_2AI[\exp(-q_2L)]}{(D_2q_2A-VK_1s)\exp(-2q_2L)+(D_2q_2A+VK_1s)} \qquad \textbf{(12)}$$

where

$$q_i = \sqrt{\frac{K_i}{D_i}(s+\lambda)} \qquad \textbf{(13)}$$

and s is the Laplace transform variable. Evaluation of the release using Equation (11) or (12) will be conservative because the mass transfer resistances of the media inside and outside the pinhole are assumed to be negligible.

2.3 MASS TRANSPORT THROUGH THE PINHOLE FOR A CONSTANT CONCENTRATION SOURCE

A constant concentration source refers to a condition where the concentration of a radionuclide or other aqueous species is maintained at a constant value inside the container for the duration of the assessment. Such a condition would occur when a radionuclide or other aqueous species precipitates so that its concentration in solution is determined by its solubility. For constant concentration, the boundary condition at x=0 becomes:

$$C(0,t) = C_o. \qquad \textbf{(14)}$$

The solution for the mass transfer rate exiting the pinhole for constant concentration, $G_2(t)$, obtained by standard methods (Carslaw and Jaeger 1959), can be expressed as the sum of a steady state expression, G_2^s, given by the first term of Equation (15) and a transient series expressed as a summation over n:

$$G_2(t) = \frac{D_2C_oAg_2}{\sinh(g_2L)} + 2\sum_{n=1}^{\infty} \frac{(-1)^n a_2 D_2 C_o A \exp[-(a_n+\lambda)t]}{(a_n+\lambda)L} \qquad \textbf{(15)}$$

where

$$g_i = \sqrt{\frac{K_i\lambda}{D_i}} \qquad (16)$$

and

$$a_n = \frac{(n\pi)^2 D_2}{K_2 L^2}. \qquad (17)$$

In the limit as λ goes to zero, $G^s_2 = D_2 C_o A/L$.

Alternatively, $G_2(t)$ can be found by numerically inverting the Laplace transform solution (Talbot 1979), $G^{\mathcal{L}}_2(s)$ where

$$G^{\mathcal{L}}_2(s) = \frac{D_2 C_o A q_2}{\sinh(q_2 L)\, s} \qquad (18)$$

and s is the Laplace transform variable.

3. MASS TRANSPORT OUTSIDE THE PINHOLE (MEDIUM 3)

3.1 MASS TRANSPORT OUTSIDE THE PINHOLE FOR AN INVENTORY-LIMITED SOURCE

To determine mass transport outside the pinhole, it is assumed that the container dimensions are large enough to be considered infinite in relation to the pinhole radius and that the external container surface can be considered as a plane surface at x=0. The mass transfer resistance outside the container is assumed to be large enough that medium 1 and medium 2 can be neglected. The mass balance equation in three dimensions in Cartesian coordinates, x,y and z is:

$$K_3 \frac{\partial C}{\partial t} = D_3 \left(\frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2} + \frac{\partial^2 C}{\partial z^2} \right) - \lambda\, K_3 C \qquad (19)$$

where K_3 is the capacity factor of the medium outside the pinhole and D_3 is the total intrinsic diffusion coefficient outside the pinhole.

The origin of the coordinate system is chosen to be at the centre of the pinhole at the outer container surface. Initially the concentration of radionuclide in the medium external to the container is assumed to be zero. The Laplace transform of the Green's function, $G(x,y,z,s|x',y',z')$ (Chambré et al. 1986, Carslaw and Jaeger 1959) for the half space, $x > 0$, having a zero flux boundary condition at $x=0$ and satisfying the mass balance equation and initial condition is:

$$G(x,y,z,s|x',y',z') = \frac{\exp\left(-q_3\sqrt{(x-x')^2+(y-y')^2+(z-z')^2}\right)}{2\pi D_3\sqrt{(x-x')^2+(y-y')^2+(z-z')^2}} \qquad (20)$$

where q_3 is given by Equation (13) with the subscript 3 indicating the medium outside the container and x',y',z' are the coordinates of the point source of unit strength associated with the Green's function.

Referring to Equations (4) and (6) and assuming that the Laplace transform of the outward flux, $S(s)$, is constant over the cross section of the pinhole, the Laplace transform of the boundary condition at $x=0$ for an inventory-limited source, can be expressed as:

$$K_1 VCs = I - \lambda K_1 VC - S(s)A. \qquad (21)$$

According to the theory of Green's functions, the Laplace transform of the concentration over the cross section of the pinhole, can be expressed as:

$$C(0,y,z,s) = S(s)\iint_{A(y',z')} G(0,y,z,s|0,y',z')\,dA \qquad (22)$$

where dA is an element of area on the cross section of the pinhole and $A(y',z')$ is the area of the pinhole. Carslaw and Jaeger (1959) have evaluated the integral in Equation (22) for a circle of radius, R. From Carslaw and Jaeger (Carslaw and Jaeger 1959, page 260), the Laplace transform of the concentration at the centre of the hole can be expressed as:

$$C(0,0,0,s) = \frac{S(s)\,[1-\exp(-Rq_3)]}{D_3 q_3} \qquad (23)$$

The Laplace transform of the total mass transfer rate, $F_3^{\mathcal{A}}(s) = S(s)A$, over the cross section of a circular hole, can be determined by applying the concentration at the centre to the entire cross section of the hole, and combining the result with Equation (21) to yield:

$$F_3^{\mathcal{A}}(s) = \frac{IK_3 A}{K_1 Vq_3\,[1-\exp(-q_3 R)] + K_3 A}. \qquad (24)$$

An approximate expression for the mass transfer rate from the hole, $F_3(t)$, can be obtained by expanding the exponential term in Equation (24) in a series, retaining only the first two terms of the series and using a table of Laplace transforms (Healy 1967) to obtain:

$$F_3(t) = \frac{IAD_3}{K_1 VR} \exp\left(-\frac{AD_3}{K_1 VR} t - \lambda t\right), \qquad t > \frac{100 K_3 R^2}{D_3}. \tag{25}$$

Equation (25) applies for times when the diffusion front has progressed a distance of more than ten radii (of the pinhole) into medium 3 ($t > 100K_3R^2/D_3$). For these times, sorption in the external medium no longer has a significant effect on the concentration gradient at the exit of the pinhole such that the mass transfer rates from the pinhole become independent of K_3.

Chambré et al. (1986) derived ratios between the mass transfer rates from circles and ellipses of varying eccentricity having the same cross sectional area. For steady state conditions the dependence of the mass transfer rates on the shape of the pinhole cross section is not large (less than a factor of 2 for elliptical holes with semi-axes ratios greater than 0.1). Thus it is reasonable to assume that the shape of the pinhole is not important for most practical purposes.

3.2 MASS TRANSPORT OUTSIDE THE CONTAINER FOR CONSTANT CONCENTRATION

For constant concentration, the Laplace transform of the boundary condition at $x=0$ is:

$$C(0,s) = \frac{C_o}{s}. \tag{26}$$

Applying the method described for the inventory-limited release, the Laplace transform of the total mass transfer rate from the hole, $G^{\mathcal{L}}_3(s)$, can be expressed as:

$$G_3^{\mathcal{L}} = \frac{\pi R^2 D_3 C_o q_3}{s[1-\exp[-q_3 R)]}. \tag{27}$$

The steady state mass transfer rate in the time domain, G_3^s, can be found by multiplying Equation (27) by s and allowing s to go to zero to obtain:

$$G_3^s = \frac{\pi R^2 D_3 C_o g_3}{1-\exp(-g_3 R)} \tag{28}$$

where g_3 is given by Equation (16).

In the limit as λ goes to zero, $G_3{}^s$ becomes $\pi D_3 C_o R$. This can be compared with the expression, $4D_3C_oR$, from Rae (1985) for the same problem. The small difference in the initial constant (π rather than 4) is due to the assumptions used here, that the flux and concentration are uniform over the cross sectional surface area of the hole whereas Rae allowed the flux and concentration to vary.

The inversion of Equation (27) and the steady state mass transfer rate, $4D_3C_oR$, from Rae (1985) are illustrated in Figure 2 for the representative parameter values given in Table 1.

TABLE 1

REPRESENTATIVE PARAMETER VALUES

description	symbol	values	units
diffusion coefficient in medium 3	D_3	$3\text{x}10^{-4}$	m^2a^{-1}
area of pinhole	A	$1\text{x}10^{-6}$	m^2
capacity factor in medium 1	K_1	0.3	none
capacity factor in medium 3	K_3	0.3	none
internal volume of container	V	0.5	m^3
radionuclide decay constant	λ	0.0	a^{-1}
constant concentration	C_o	1.0	mol/m^3
initial inventory	I	1.0	mol

The solution from Rae, for a zero radioactive decay constant, gives higher mass transfer rates than Equation (27) except at very early times. The difference between the two steady state mass transfer rates shown in Figure 2 is not large (less than a factor of 1.3) and would normally be less than the variability associated with the uncertainty in parameter values. According to Equation (27), the mass transfer rate increases as the decay constant increases, an effect that is somewhat counter-intuitive. As the decay constant increases, the depletion of the concentration outside the pinhole is increased. This depletion causes the concentration gradient to increase. It is assumed, however, that the concentration at the exit from the pinhole is maintained at a constant concentration. The combination of these effects results in increased mass transfer rates from the pinhole for increasing decay constants. To ensure conservatism, Equations (27) and (28) can be used for all values of the decay constant with the constant π replaced by 4.

AECL EACL
Memo Note

WASTE TECHNOLOGY DIVISION
Environmental and Safety Assessment Branch
Fax: 204-753-2690

File No. R-3146
ESAB-96-094

1996 April 11

TO: J. Hoffmann

FROM: D.M. LeNeveu

<u>ERRATUM FOR AECL-11540 COG-96-68</u>

Please find attached an erratum for AECL-11540, COG-96-68.

DML/eh

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ERRATUM

Equation (12) on page 6 now reads:

$$F_2(L,s)=\frac{2D_2q_2AI[\exp(-q_2L)]}{(D_2q_2A-VK_1s)\exp(-2q_2L)+(D_2q_2A+VK_1s)} \qquad (12)$$

Equation (12) should read:

$$F_2(L,s)=\frac{2D_2q_2AI\exp(-q_2L)}{[D_2q_2A-VK_1(s+\lambda)]\exp(-2q_2L)+[D_2q_2A+VK_1(s+\lambda)]} \qquad (12)$$

Figure 2 now has Eqn(25) in the legend. Figure 2 should have Eqn(27)in the legend.

Figure 3 now has Eqn(22), Eqn(23) and Eqn(28) in the legend. Figure 3 should have Eqn(24), Eqn(25) and Eqn(30) in the legend.

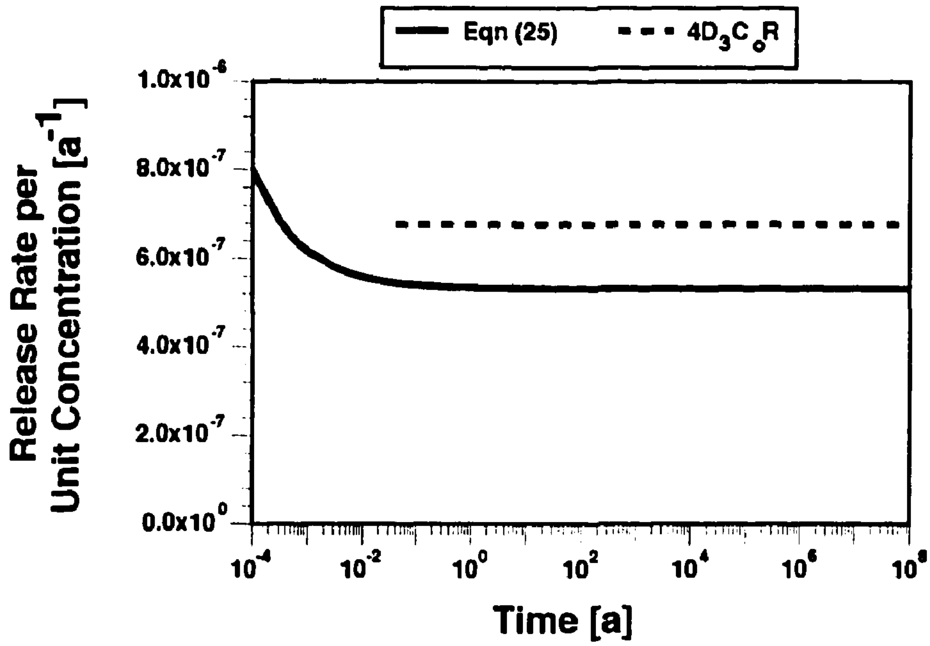

FIGURE 2: Release Rates for Constant Concentration

3.3 INVENTORY-LIMITED RELEASE DETERMINED FROM PSEUDO-STEADY STATE CONDITIONS

In the inventory-limited case, if it is assumed that the concentration inside the container changes slowly enough that pseudo-steady state conditions are achieved at all times, then, using the steady state expression from Rae, the Laplace transform of the relationship between total mass transfer rate from the hole and concentration at the exit of the pinhole can be expressed as:

$$F_3^{\mathcal{L}}(s) = 4D_3RC(s) \qquad \textbf{(29)}$$

Equation (29) and the equation for the boundary condition at x=0 (Equation (21)) can be used to solve for $F^{\mathcal{L}}_3(s)$ which can be inverted from a table of transforms (Healy 1967) to obtain:

$$F_3(t) = \frac{4ID_3R}{K_1V}\exp\left(-\frac{4D_3R}{K_1V}t - \lambda\ t\right), \qquad t > \frac{100K_3R^2}{D_3}. \qquad \textbf{(30)}$$

Equation (25) and Equation (30) for $F_3(t)$ differ only by the constants 4 and π.

3.4 PREFERRED FORMULATION FOR INVENTORY-LIMITED RELEASE OUTSIDE THE CONTAINER

The question remains, which equation should be used to represent the mass transfer rate from the pinhole for an inventory-limited source, Equation (24), (25) or (30)? The mass transfer rates from the pinhole for these three equations are compared in Figure 3 for representative parameter values given in Table 1. As shown in Figure 3, the inversion of Equation (24) gives the highest values of the mass transfer rate at very early times (less than one year). Afterwards, until very long times, Equation (30) gives the highest mass transfer rates. At very long times, (greater than 4×10^5 years) the inversion of Equation (24) gives slightly higher values of mass transfer rate. In waste management studies, the equation that predicts the highest mass transfer rates occurring the earliest is usually used. The early highest mass transfer rates from Equation (24) occur over such a small time interval that the contribution to the overall release of radionuclides is insignificant. A more salient factor is that the early high release is an artifact of modelling only one medium. In reality, it would take some time for the container to fill with water and for the concentration to move through the pinhole such that at early times, the mass transfer rate would be rising from a zero value rather than decreasing from a high value.

From these considerations it would be preferable to use Equation (30), over the entire time domain, for determining mass transfer rates in the external medium. Careful experimentation could perhaps determine the most accurate formulation; however, the

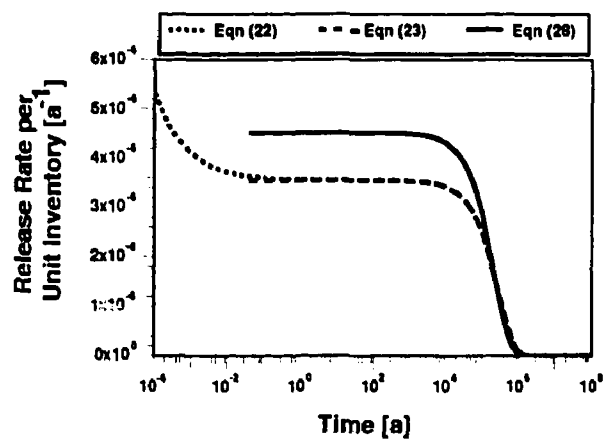

FIGURE 3: Release Rates for an Inventory-Limited Source

uncertainty in the parameter values might be too large to allow for discrimination between the small differences obtained through the use of the different equations.

4. MASS TRANSFER RESISTANCE

The medium with the largest mass transfer resistance will have the largest influence in determining the overall mass transfer rates from a multi-media system (Rae 1985, Pescatore and Sastre 1988). This means that the equations in this study, that deal with mass transport in a single medium, can be used to approximate the entire system provided that the medium with the largest mass transfer resistance is identified. The addition of mass transfer resistances can also be used to determine expressions for mass transfer rates that take into account mass transport in all the media in the system simultaneously under steady state or pseudo- steady state conditions (Crank 1956).

By comparing the equations for $F_2(L,t)$ and $F_3(t)$ and the equations for G^s_2 and G^s_3 for the case where the decay constant is zero, the mass transfer resistances for the pinhole and for the external medium are defined to be L/D_2 and R/D_3 respectively. These are consistent with the definitions from standard sources (Crank 1956). For the constant concentration case, when the decay constants are non-zero, the standard definitions for mass transfer resistance do not apply. For this case, it is recommended that the medium with the smallest steady state mass transfer rate from Equations (15) and (27), be considered to be the most restrictive and to have the largest mass transfer resistance.

The work of DePaoli and Scott (1993) indicates the importance of including transient conditions in determining which medium dominates in controlling mass transport. However, because the dimensions of the pinhole are small, transients should be of short duration inside the pinhole and at the exit to the pinhole compared to the duration of the transient through the complete extent of the medium external to the pinhole. In this study it is suggested that the mass transfer resistance based on steady state conditions be used to determine the single most restrictive medium that controls mass transport. Use of a single medium for mass transport always results in a conservative determination of mass transfer rates because the mass transfer resistances of the other media are neglected.

Nevertheless, addition of mass transfer resistances can be used for the simultaneous consideration more than one medium. By examining the form of Equations (11) and (25) for the inventory-limited source, the total resistance R_T of the two medium 2 and medium 3 can be expressed as:

$$R_T = \frac{L}{D_2} + \frac{R}{D_3} \qquad (31)$$

Using the total resistance, the mass transfer rate from the pinhole for the inventory-limited case, $F(t)$, can be expressed as:

$$F(t) = \frac{IA}{K_1 V R_T} \exp\left(-\frac{A}{K_1 V R_T} t - \lambda t\right), \quad \frac{K_2 A L}{K_1 V} \ll 1 . \qquad (32)$$

Equation (32) would apply when the concentration was changing slowly enough that pseudo-steady state conditions would apply in both media, ($t > 100K_3R^2/D_3 + K_2L^2/D_2$).

By analogy with medium 3, the mass transfer resistance for medium 1 would be, R/D_1. The resistance of medium 1, could be added to the total resistance such that Equation (32) would apply simultaneously to all media in the system provided that an additional factor of $100K_1R^2/D_1$ is added to account for the increase in the duration of the transient. A similar result can be obtained for the constant concentration case under steady state conditions when the decay constant is zero.

5. MULTIPLE HOLES

In general, the equations in this paper should only be used when the area of the pinhole is very much smaller than the surface area of the container and when multiple holes are far enough apart that they can be treated independently. Pescatore and Sastre (1988) described the conditions under which multiple holes become close enough to interact.

6. CONCLUSION

For most analyses of the containment of radioactive wastes, radionuclide mass transfer rates from pinholes in containers can be adequately modelled by the equations developed here. The medium with the largest mass transfer resistance should be used for the equations with mass transport in a single medium. This will provide a conservative determination of the mass transfer rate from pinholes because the mass transfer resistances of the other media have been neglected. The equations give both transient and steady state mass transfer rates from a pinhole for constant concentration source conditions and for inventory-limited source conditions, and take into account the effect of radioactive decay. The concept of addition of resistances has been used to obtain an expression for inventory-limited release under pseudo-steady state conditions that takes into account mass transport in all media in the system simultaneously.

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