United States
Environmental Protection
Agency

Office of
Radiation Programs
Washington DC 20460

EPA 520/8-81-007
1981

Radiation

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June 1981

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ABSTRACT

The short- and long-term leach rates of radionuclides for the three-dimensional diffusive flow and for purely axial diffusive flow from a finite cylinder of solidified waste are determined here. These analytical results are compared with the ones obtained numerically by Hung (Hu80) for purely axial flow.

CONTENTS

1. PURELY AXIAL FLOW

1.1 Introduction

A common method for the disposal of radioactive waste is to embed the radionuclides into a solid container. However, these particles can still escape from the container and thus be a hazard to the environment.

The short- and long-term leach rates of radionuclides for the diffusive flow from a finite cylinder of solidified waste are discussed here. Their transport is governed by diffusion, desorption, and radioactive decay, and is assumed to be in one direction only. (For example, the system presented here models a cylindrical container of length L, where all the faces, except for one end, are insulated).

This same system was discussed by Hung (Hu80). The solution to the basic differential equation with prescribed boundary conditions was obtained by numerical analysis. However, the equation has a fairly simple solution, which will be discussed below. Comparisions of these results will be made with those of Hung, and also to the solution for a semi-infinite cylinder with insulated sides.

1.2 Solution to the Model Equation

The basic equation for the transport of radionuclides through the porous medium of the container (which has been immersed in an aqueous solution), where diffusion, desorption, and radioactive decay are present, is given by Hung (Hu80) as

$$\frac{\partial c}{\partial t} = \frac{D_e}{\epsilon R} \frac{\partial^2 c}{\partial x^2} - \lambda_d c , \quad 0 \le x \le L , \tag{1.1}$$

where c is the concentration of radionuclides (Ci/cm^3) in the aqueous solution which is saturating the waste solid, D_e is the effective diffusivity through the solidified waste ($cm^2/year$), ϵ is the porosity, R is the retardation factor defined in (Hu80), and λ_d is the decay constant for the radionuclide.

The initial and boundary conditions on the container are

$$c(x,0) = C_0 \qquad 0 < x < L, \tag{1.2}$$

$$c(0,t) = 0 \qquad t > 0, \tag{1.3}$$

$$\frac{\partial c}{\partial x}(L,t) = 0 \qquad t > 0 . \tag{1.4}$$

Therefore, it is assumed that the equilibrium state of desorption has been reached at time t=0, there is no accumulation of radionuclides at the end x=0 and that the other end, x=L, is insulated.

The equation (1.1) with conditions (1.2)-(1.4) can be solved by the standard techniques of separation of variables and the superposition of normal modes (Hi76). This gives

$$c(x,t)=\frac{4C_o}{\pi} e^{-\lambda_d t} \sum_{n=0}^{\infty} \frac{1}{2n+1} \sin \frac{(2n+1)\pi x}{2L} e^{-\left[\frac{(2n+1)\pi}{2L}\right]^2 \kappa t} , \qquad (1.5)$$

where $\kappa = D_e/\epsilon R$.

Now that the radionuclide concentration in the aqueous solution, which saturates the solidified waste, is known, the rate of leaching l(t) can be obtained from

$$l(t) = D_e A_r \frac{\partial c}{\partial x}(0,t) \qquad \text{(Ci/year)}, \qquad (1.6)$$

where A_r is the cross-sectional area of the solid (cm^2). It is often useful to determine the rate of leaching as a fraction of the total initial activity A_o. Here

$$A_o = \epsilon R C_o A_r L \quad ; \qquad (1.7)$$

then from (1.5)-(1.7), the rate of radionuclide leaching as a fraction of the initial activity per year is

$$\frac{l(t)}{A_o} = 2L_c e^{-\lambda_d t} \sum_{n=0}^{\infty} e^{-\left[\frac{(2n+1)\pi}{2}\right]^2 L_c t} . \qquad (1.8)$$

Here a new parameter, the leaching number, L_c, has been defined:

$$L_c = \frac{\kappa}{L^2} = \frac{D_e}{\epsilon R L^2} . \qquad (1.9)$$

In order to conform to Hung's notation, we introduce a dimensionless time variable t', a dimensionless leaching parameter l' and a leaching number L_n:

$$t' = \frac{D_e}{\epsilon R L^2} t = L_c t ,$$

$$l' = \frac{\epsilon R L^2}{A_o D_e} l = \frac{l}{A_o L_c} , \quad (1.10)$$

and $L_n = \lambda_d / L_c$.

Substitution of (1.10) into (1.8) gives

$$l' = 2e^{-L_n t'} \sum_{n=0}^{\infty} e^{-[\frac{(2n+1)\pi}{2}]^2 t'} . \quad (1.11)$$

In the following two sections we consider approximations to l' for t' small and for t' sufficiently large (near one or greater).

1.3 Short-Time Leach Rates

An estimate of the dimensionless leach rate l', given by (1.11) is now required. This involves making an asymptotic approximation to the summation S,

$$S = \sum_{n=0}^{\infty} e^{-[\frac{(2n+1)\pi}{2}]^2 t'} \quad (1.12)$$

for $0 < t' << 1$. To do this, compare the summation with the area under the graph of $e^{-[\frac{n\pi}{2}]^2 t'}$ for $0 \leq n \leq \infty$; this is illustrated in Figure 1. Since

$$\int_0^\infty e^{-[\frac{n\pi}{2}]^2 t'} \, dn = \frac{1}{\sqrt{\pi t'}} , \tag{1.13}$$

we have, from consideration of Figure 1, the two unequalities

$$S < \frac{1}{2} \left[\frac{1}{\sqrt{\pi t'}} + e^{-\frac{\pi^2}{4} t'}\right] < \frac{1}{2} \left[\frac{1}{\sqrt{\pi t'}} + 1\right] , \tag{1.14}$$

and

$$S > \frac{1}{2} \left[\frac{1}{\sqrt{\pi t'}} - 1\right] , \tag{1.15}$$

for all values of $t' > 0$. Therefore for $0 < t' << 1$

$$S \sim \frac{1}{2 \sqrt{\pi t'}} . \tag{1.16}$$

Combining the result with equation (1.11), and assuming that $L_n t' << 1$, as well as $t' << 1$,

$$1' \sim \frac{1}{\sqrt{\pi t'}} . \tag{1.17}$$

Hence, we conclude that the short-time (dimensionless) leach rate for the finite-dimensional geometry is the same as that for the semi-infinite geometry (as shown in Appendix A). This confirms the results of Hung (in his Figures 2 and 3).

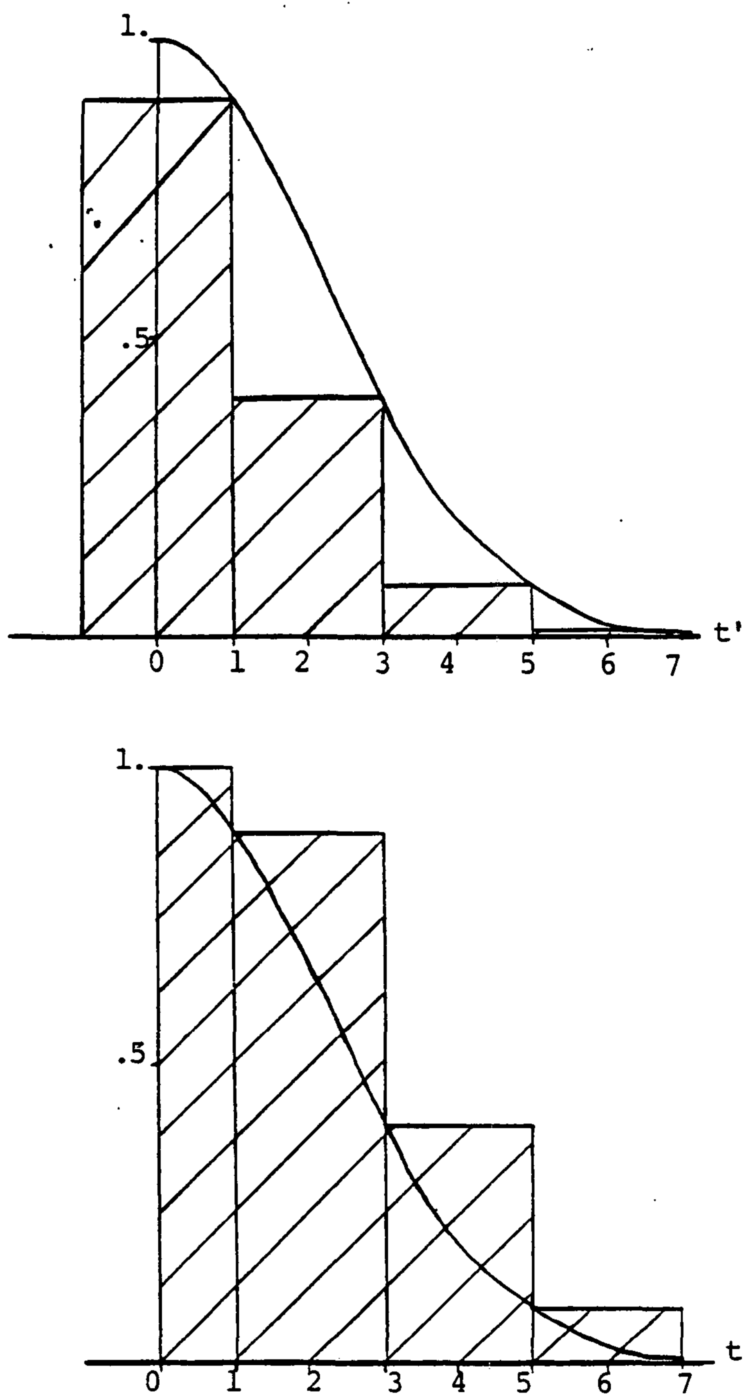

Figure 1. Graphs of $e^{-(n\pi/2)^2 t'}$ versus n for $t' > 0$

1.4 Long-Time Leach Rates

For sufficiently large values of (dimensionless) time, the value of l' can be approximated by the first term in the series:

$$l' \sim 2 e^{-[L_n + \frac{\pi^2}{4}] t'} \quad . \tag{1.18}$$

For intermediate values of time, then the first few terms of the series expansion (1.11) will be sufficient to give a good approximation to l'.

The values given by (1.18), for t' near one, match well with the values illustrated in Figure 3 by Hung, which were obtained by numerically solving the set of difference equations associated with equation (1.1).

1.5 Conclusions

The leach rates for the finite-dimensional model system can be well approximated by the simple formulas:

$$l' \sim \frac{1}{\sqrt{\pi t'}} \qquad \text{for } t' << 1 \ , \tag{1.19}$$

and

$$l' \sim 2 e^{-[L_n + \frac{\pi^2}{4}] t'} \qquad \text{for } t' \text{ sufficiently large,} \tag{1.20}$$

for the dimensionless variables l' and t'. These results match well with those of Hung, which were carried out more expensively by using the computer to solve the basic equation.

APPENDIX A

The analagous problem to the one discussed in Section 2, but for $0 \leq x < \infty$ is

$$\frac{\partial c}{\partial t} = \frac{\partial^2 c}{\partial x^2} - \lambda\, c_d \ ,$$

$$\begin{aligned} c(x,0) &= C_0 \qquad x > 0\ , \\ c(0,t) &= 0 \qquad t > 0\ , \end{aligned} \tag{A.1}$$

where $\kappa = D_e/\epsilon R$. This has an exact solution

$$c(x,t) = C_0\, e^{-\lambda_d t}\ \mathrm{erf}\left(\frac{x}{2\sqrt{\kappa t}}\right) \ , \tag{A.2}$$

where erf(y) is the well-known error function

$$\mathrm{erf}(y) = \frac{2}{\sqrt{\pi}} \int_0^y e^{-u^2}\, du \ . \tag{A.3}$$

Then the leach rate from the face x=0 is just

$$l = D_e A_r \frac{\partial c}{\partial x}(0,t) = D_e A_r C_0 \frac{e^{-\lambda_d t}}{\sqrt{\pi \kappa t}} \ . \tag{A.4}$$

Using the notation of Section 1.2, this gives

$$I' = \frac{I}{A_o L_c} = \frac{e^{-L_n t'}}{\sqrt{\pi t'}} \quad . \tag{A.5}$$

For small t' and $L_n t'$, this may be approximated as

$$I' = \frac{1}{\sqrt{\pi t'}} \quad . \tag{A.6}$$

2. THREE-DIMENSIONAL FLOW

2.1 Introduction

Numerous studies have been made to determine the leach rates of radionuclides from solid waste. However, the results obtained so far are of limited use in practical applications because the basic assumptions are often not physically acceptable.

For modelling purposes, it is usually assumed that the flow out of the solidified waste is in one direction only. Both the semi-infinite (Go74) and finite-dimensional (Hu80) models, with uni-directional flow, have been studied. However, these geometries are too simplistic to give reliable results. A real canister is usually cylindrical and it cannot be assumed that all of its exterior surfaces, except for one end face, are insulated. In fact, there will be a flux out of all the three faces of the cylinder, giving rise to a three-dimensional flow.

This paper addresses the problem of short- and long-time leach rates out of a cylinder, where the radionuclides can escape out of all the sides. Some of the results will be compared with Hung's (Hu80). He used a "scaling-effect" relationship, without adequate justification, to approximate the three-dimensional flow from a cylinder with the one-dimensional flow. However, we will show that his results are not necessarily accurate.

The other assumptions often made are that the chemical and physical characteristics of the solidified waste stay the same over long time periods. This may also be simplistic.

2.2 Determination of the Leach Rate

The basic equation for the transport of radionuclides through the porous medium of the container (which has been immersed in an aqueous solution), where diffusion, desorption, and radioactive decay are present, is given by Hung (Hu80). For a cylindrical geometry, with axial symmetry, this is most conveniently given in cylindrical co-ordinates:

$$\frac{\partial c}{\partial t} = \frac{D_e}{\varepsilon R}\left[\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial c}{\partial r}\right) + \frac{\partial^2 c}{\partial z^2}\right] - \lambda_d c \, , \qquad (2.1)$$

where r and z are the radial and axial co-ordinates, respectively. Here c is the concentration of radionuclides in the aqueous solution which saturates the solid waste, D_e is the effective diffusivity, ε is the porosity, R is the retardation factor defined in (Hu80), and λ_d is the decay constant for the radionuclide.

The initial and boundary conditions for the container are:

$$c = C_o \qquad t = 0,\ 0 \leq r < a,\ 0 < z < L, \tag{2.2}$$

$$c = 0 \qquad z = 0,\ z = L,\ 0 \leq r < a,\ t > 0, \tag{2.3}$$

$$c = 0 \qquad r = a,\ 0 < z < L,\ t > 0. \tag{2.4}$$

Therefore, it is assumed that the equilibrium state of desorption has been reached at time $t = 0$ and that there is no accumulation of radionuclides on the exterior surfaces of the container.

The equation (2.1) with boundary conditions (2.3) - (2.4) can be solved by the standard techniques of separation of variables and the superposition of normal modes as

$$c(r,z,t) = e^{-\lambda_d t} \sum_{m=1}^{\infty} \sum_{n=1}^{\infty} B_{mn} J_0(\mu_m r) \sin \frac{n\pi z}{L}\, e^{-[(\frac{n\pi}{L})^2 + \mu_m^2]\kappa t}, \tag{2.5}$$

where $\kappa = D_e/\varepsilon R$. The $\mu_m a$ are the set of positive zeros of $J_0(x)$ and the B_{mn} are constants which are determined by the initial condition (2.2) as

$$B_{mn} = \frac{4C_o}{a^2 L\, [J_1(\mu_m a)]^2} \int_0^a r J_0(\mu_m r)\, dr \int_0^L \sin \frac{n\pi z}{L}\, dz\ . \tag{2.6}$$

Here the orthogonality relations for the $J_0(\mu_m r)$ and $\sin \frac{n\pi z}{L}$ functions are used (Hi76)]. Evaluation of the integrals gives

$$B_{mn} = \begin{cases} \dfrac{8C_o}{\pi a n \mu_m J_1(\mu_m a)} & n \text{ odd} \\ 0 & n \text{ even} \end{cases} \qquad (2.7)$$

Substitution of (2.7) into (2.5) then gives the solution as

$$c(r,z,t) = \frac{8C_o}{\pi a} e^{-\lambda_d t} \sum_{\substack{m,n=1 \\ n \text{ odd}}}^{\infty} \frac{J_o(\mu_m r)}{n\mu_m J_1(\mu_m a)} \sin \frac{n\pi z}{L} e^{-[(\frac{n\pi}{L})+\mu_m^2]\kappa t} \qquad (2.8)$$

Now that the radionuclide concentration is known, the rate of leaching l(t) can be obtained. Since the flux of radionuclides, $\underset{\sim}{J}$, is given by

$$\underset{\sim}{J} = - D_e \text{ grad } c = -D_e\left(\frac{\partial c}{\partial r} \hat{r} + \frac{\partial c}{\partial z} \hat{z}\right) \quad , \qquad (2.9)$$

the concentration of radionuclides which passes out of the cylinder per unit time is

$$l(t) = -\iint_S \underset{\sim}{J} \cdot \hat{n} \; dS \quad , \qquad (2.10)$$

where $\hat{n}$ is the unit normal of the surface in the outward direction and the integral is taken over the whole surface. Therefore, the integral in (2.10) has three parts - one corresponding to the end face z = 0, a similar one for z = L, and one for the surface r = a. Writing this out more fully we must evaluate

$$I(t) = \int_0^{2\pi}\int_0^a D_e \left.\frac{\partial c}{\partial z}\right|_{z=0} r\,dr\,d\theta - \int_0^{2\pi}\int_0^a D_e \left.\frac{\partial c}{\partial z}\right|_{z=L} r\,dr\,d\theta - \int_0^{2\pi}\int_0^L D_e \left.\frac{\partial c}{\partial r}\right|_{r=a} a\,dz\,d\theta \quad (2.11)$$

From equation (2.8), it is easily verified that

$$I(t) = \frac{32}{L} D_e \pi C_o e^{-\lambda_d t} \sum_{\substack{m,n=1 \\ n\ odd}}^{\infty} \left[\frac{1}{(\frac{n\pi}{L})^2} + \frac{1}{\mu_m^2} \right] e^{-[(\frac{n\pi}{L})^2 + \mu_m^2]\kappa t} . \quad (2.12)$$

It is often useful to determine the rate of leaching as a fraction of the total initial activity A_o. Here

$$A_o = \epsilon R C_o \pi a^2 L , \quad (2.13)$$

so that

$$\frac{I(t)}{A_o} = \frac{32\kappa}{a^2 L^2} e^{-\lambda_d t} \sum_{\substack{m,n=1 \\ n\ odd}}^{\infty} \left[\frac{1}{(\frac{n\pi}{L})^2} + \frac{1}{\mu_m^2} \right] e^{-[(\frac{n\pi}{L})^2 + \mu_m^2]\kappa t} . \quad (2.14)$$

In the following two sections we consider approximations to $I(t)/A_o$ for small and long periods of time and compare our results to Hung's for a particular example he discusses.

2.3 Short-Time Leach Rates

An estimate of the rate of radionuclide leaching as a fraction of the initial activity, given by equation (2.14) is now required. This involves making an asymptotic approximation to the double summation

$$S = \sum_{\substack{m,n=1 \\ n \text{ odd}}}^{\infty} \left[\frac{1}{(\frac{n\pi}{L})^2} + \frac{1}{\mu_m^2} \right] e^{-[(\frac{n\pi}{L})^2 + \mu_m^2]\kappa t} , \tag{2.15}$$

for t sufficiently small. With some manipulation this may be rewritten as

$$S = \sum_{\substack{n=1 \\ n \text{ odd}}}^{\infty} e^{-(\frac{n\pi}{L})^2 \kappa t} \sum_{m=1}^{\infty} \frac{1}{\mu_m^2} e^{-\mu_m^2 \kappa t}$$

$$+ \sum_{m=1}^{\infty} e^{-\mu_m^2 \kappa t} \sum_{\substack{n=1 \\ n \text{ odd}}}^{\infty} \frac{1}{(\frac{n\pi}{L})^2} e^{-(\frac{n\pi}{L})^2 \kappa t} . \tag{2.16}$$

It is convenient to define two dimensionless time scales

$$\tau = \frac{\kappa t}{L^2} \quad \text{and} \quad T = \frac{\kappa t}{a^2} . \tag{2.17}$$

In Appendix B, the asymptotic forms of each of the series in equation (2.16) is determined for small values of τ and T. The results there give

$$S \sim \frac{1}{4\sqrt{\pi\tau}} \cdot \frac{a^2}{4} + \frac{1}{2\sqrt{\pi T}} \cdot \frac{L^2}{8} , \quad \tau, T \ll 1$$
$$- \frac{1}{16\sqrt{\pi}} \left[\frac{a^2}{\sqrt{\tau}} + \frac{L^2}{\sqrt{T}} \right] . \tag{2.18}$$

Therefore, for small values of $\kappa t/L^2$, $\kappa t/a^2$ and $\lambda_d t$, the rate of leaching as a fraction of the initial activity is

$$\frac{l(t)}{A_o} \simeq \frac{2\kappa}{\sqrt{\pi}} \left[\frac{1}{a^2\sqrt{T}} + \frac{1}{L^2\sqrt{\tau}}\right] \simeq 2\sqrt{\frac{\kappa}{\pi t}} \left[\frac{1}{a} + \frac{1}{L}\right] \quad . \tag{2.19}$$

This asymptotic form shows that the rate of leaching for the three-dimensional flow from a cylinder is made up of two contributions--one that would occur from the purely radial flow and the other from the purely axial flow.

Indeed, since the radius a of a canister used for disposal purposes is usually smaller than the length L, choosing a time t such that $\kappa t/a^2 << 1$ shows us that the dominant contribution to the leach rate is from the radial flow.

This shows that the one-dimensional axial flow from a cylinder will not accurately determine the short time leach rates from a cylinder, since the radial flow will usually contribute a larger amount.

If τ and T are small, but the decay constant is large enough so that $\lambda_d t$ is no longer small, then the fractional leach rate is

$$\frac{l(t)}{A_o} \simeq 2\, e^{-\lambda_d t} \sqrt{\frac{\kappa}{\pi t}} \left[\frac{1}{a} + \frac{1}{L}\right] \quad , \tag{2.20}$$

and the above remarks remain valid. In fact, the example presented by Hung (Hu80) for the predicted leach rate at time t = 100 years is really a problem of determining the short-term leach rate for the three-dimensional flow described here. The parameters are as follows: the number κ, which measures the physical and chemical properties of the radionuclides in the solidified waste medium, can be determined from the leaching constant for the 5-cm long test piece, so that

$$\kappa = 3.74 \times 10^{-6} \times (5)^2 = 9.35 \times 10^{-5} \text{ cm}^2/\text{day}. \quad (2.21)$$

Also

$$\begin{aligned} \lambda_d &= 6.33 \times 10^{-5}/\text{day} \\ a &= 30 \text{ cm} \\ L &= 90 \text{ cm} \\ t &= 3.65 \times 10^4 \text{ day}. \end{aligned} \quad (2.22)$$

Therefore

$$T = \frac{\kappa t}{a^2} = 3.79 \times 10^{-3} > \frac{\kappa t}{L^2} , \quad (2.23)$$

hence both the dimensionless time scales τ and T are small. However,

$$\lambda_d t = 2.31, \quad (2.24)$$

so that the short term leach rate as given by equation (2.20) applies. This gives

$$\frac{l(t)}{A_0} = 2.51 \times 10^{-7}/\text{day}; \quad (2.25)$$

so that, for this particular choice of the parameters, the predicted fractional leach rate has the same order of magnitude as that obtained by Hung (his equation (32)), with the use of his scaling effect argument. However, this will not always be true, the relative size of the parameters is important.

2.4 Long-Term Leach Rates

For sufficiently large values of the dimensionless time variable $\kappa t/a^2$ and $\kappa t/L^2$, the value of $l(t)/A_0$ can be approximated by the first term in the series (2.14)

$$\frac{l(t)}{A_0} \sim \frac{32\, e^{-\lambda_d t}}{a^2 L^2} \left[\frac{L^2}{\pi^2} + \frac{a^2}{\xi_1^2}\right] e^{-\left(\frac{\pi^2}{L^2} + \frac{\xi_1^2}{a^2}\right)\kappa t} . \quad (2.26)$$

wnere ξ_1 = 2.405 is the first positive zero of $J_0(x)$. For intermediate values of (dimensionless) time, the first few terms of the series expansion (2.14) will be sufficient to give a good approximation to $1/A_0$.

APPENDIX B

1. Consider the purely axial flow from a cylinder, radius a and length L. Suppose u(z,t) satisfies

$$\begin{aligned} \frac{\partial u}{\partial t} &= \kappa \frac{\partial^2 u}{\partial z^2}, && 0 < z < L , \\ u(z,0) &= U_o && 0 < z < L , \\ u(0,t) &= u(L,t) = 0 && t > 0 . \end{aligned} \qquad (B.1)$$

The solution to this problem, when obtained by separation of variables and superposition of the normal modes, is

$$u(z,t) = \frac{4U_o}{\pi} \sum_{\substack{n=1 \\ n \text{ odd}}}^{\infty} \frac{1}{n} \sin \frac{n\pi z}{L} \, e^{-(\frac{n\pi}{L})^2 \kappa t} . \qquad (B.2)$$

Then, using the notation defined in the body of this paper, the rate of leaching as a fraction of the initial activity is

$$\frac{l(t)}{A_o} = \frac{8\kappa}{L^2} \sum_{\substack{n=1 \\ n \text{ odd}}}^{\infty} e^{-(\frac{n\pi}{L})^2 \kappa t} . \qquad (B.3)$$

However, the series in (B.2) and (B.3) converge very slowly for small values of $\kappa t/L^2$. An alternative approach to solving (B.1) is more appropriate then. This involves using the Laplace transform (Ab72). If p is the transform variable and $\bar{f}$ denotes the Laplace transfore of f, then (B.1) becomes

$$\begin{aligned} \frac{d^2\bar{u}}{dz^2} - q^2\bar{u} &= -\frac{U_o}{\kappa} , \qquad q = \sqrt{\frac{p}{\kappa}} > 0 , \\ \bar{u} &= 0 , \quad z = 0,L . \end{aligned} \qquad (B.4)$$

The solution to (B.4) is

$$\bar{u} = \frac{U_0}{p} \left[1 - \frac{\sinh qz + \sinh q(L-z)}{\sinh qL}\right] . \quad (B.5)$$

Then,

$$\frac{1}{U_0} \bar{u}_z \Big|_{z=0} = -\frac{1}{U_0} \bar{u}_z \Big|_{z=L}$$

$$= \frac{1}{\kappa q} \left[\frac{\cosh qL - 1}{\sinh qL} \right]$$

$$= \frac{1}{\kappa q} \left[\frac{1 + e^{-2qL} - 2e^{-qL}}{1 - e^{-2qL}} \right]$$

$$= \frac{1}{\kappa q} \left[1 + e^{-2qL} - 2e^{-qL}\right] \sum_{n=0}^{\infty} e^{-2nqL} \quad (B.6)$$

$$= \frac{1}{\kappa q} \left[1 + 2 \sum_{n=1}^{\infty} (-1)^n e^{-nqL}\right] .$$

Taking the inverse transform of these standard Laplace transform functions (Ab72) gives

$$\frac{\partial u}{\partial z} \Big|_{z=0} = -\frac{\partial u}{\partial z} \Big|_{z=L} = \frac{U_0}{\sqrt{\pi \kappa t}} \left[1 + 2 \sum_{n=1}^{\infty} (-1)^n e^{-\frac{n^2 L^2}{4\kappa t}}\right] . \quad (B.7)$$

Hence,

$$\frac{I(t)}{A_0} = \frac{2}{L\sqrt{\pi \kappa t}} \left[1 + 2 \sum_{n=1}^{\infty} (-1)^n e^{-\frac{n^2 L^2}{4\kappa t}}\right] . \quad (B.8)$$

Compare this with (B.3); then the following asymptotic approximation is valid for small values of $\kappa t/L^2 = \tau$:

$$\sum_{\substack{n=1 \\ n \text{ odd}}}^{\infty} e^{-n^2 \pi^2 \tau} \sim \frac{1}{4\sqrt{\pi \tau}} . \quad (B.9)$$

Similarly, from (B.2) we can evaluate the average concentration in the cylinder as a function of time:

$$v = \frac{1}{L}\int_0^L u(z,t)dz = \frac{8U_o}{\pi^2} \sum_{\substack{n=1 \\ n \text{ odd}}}^{\infty} \frac{1}{n^2} e^{-(\frac{n\pi}{L})^2 \kappa t} \quad . \tag{B.10}$$

But from (B.5) and then (B.6),

$$\bar{v} = \frac{1}{L}\int_0^L \bar{u}(z,p)\, dz = \frac{U_o}{p}\left[1 - \frac{2}{Lq}\left(\frac{\cosh qL - 1}{\sinh qL}\right)\right]$$

$$= \frac{U_o}{p}\left[1 - \frac{2}{Lq}\left(1 + 2\sum_{n=1}^{\infty}(-1)^n e^{-nqL}\right)\right] \quad . \tag{B.11}$$

Taking the inverse transform gives

$$v = U_o - 4U_o\left(\frac{\kappa t}{L^2}\right)^{1/2}\left[\pi^{-1/2} + 2\sum_{n=1}^{\infty}(-1)^n \,\text{ierfc}\, \frac{n}{2\sqrt{\frac{\kappa t}{L^2}}}\right] \quad . \tag{B.12}$$

Again comparison of (B.10) with the above equation gives, for small values of τ ,

$$\sum_{\substack{n=1 \\ n \text{ odd}}}^{\infty} \frac{1}{n^2} e^{-n^2\pi^2\tau} \sim \frac{\pi^2}{8} \quad . \tag{B.13}$$

2. Consider the purely radial flow from a cylinder of radius a and length L. Suppose u(r,t) satisfies

$$\begin{aligned} \frac{\partial u}{\partial t} &= \kappa \frac{1}{r} \frac{\partial}{\partial r}\left(r \frac{\partial u}{\partial r}\right) && 0 \le r < a \ , \\ u(r,0) &= U_o && 0 \le r < a \ , \\ u(a,t) &= 0 && t > 0 \ . \end{aligned} \tag{B.14}$$

The solution to this problem, using the separation of variable technique, is

$$u(r,t) = 2U_o \sum_{m=1}^{\infty} \frac{J_o(\xi_m r/a)}{\xi_m J_1(\xi_m)} e^{-\xi_m^2 \frac{\kappa t}{a^2}} , \qquad (B.15)$$

where $J_o(\xi_m)=0$, $\xi_m > 0$. Then,

$$\frac{I(t)}{A_o} = \frac{4\kappa}{a^2} \sum_{m=1}^{\infty} e^{-\xi_m^2 \frac{\kappa t}{a^2}} . \qquad (B.16)$$

However, the series in (B.15) and (B.16) converge very slowly for small values of $\kappa t/a^2$. The Laplace transform method is better for this case; then (B.14) becomes:

$$\frac{d^2\bar{u}}{dr^2} + \frac{1}{r}\frac{d\bar{u}}{dr} - q^2\bar{u} = -\frac{U_o}{\kappa} , \qquad q = \sqrt{\frac{p}{\kappa}} > 0, \qquad (B.17)$$

$$\bar{u} = 0, \qquad r=a .$$

The solution to (B.17) is

$$\bar{u} = \frac{U_o}{p}\left[1 - \frac{I_o(qr)}{I_o(qa)}\right] . \qquad (B.18)$$

Therefore,

$$\left.\frac{-1}{U_o}\bar{u}_r\right|_{r=a} = \frac{q}{p}\frac{I_1(qa)}{I_o(qa)} . \qquad (B.19)$$

Using the expansion of $I_v(x)$ for large values of x,

$$I_v(x) = \frac{e^x}{\sqrt{2\pi x}}\left[1 - \frac{4v^2-1}{1!\,8x} + \frac{(4v^2-1)(4v^2-3^2)}{2!\,(8x)^2} + 0\left(\frac{1}{x^3}\right)\right] + 0\left(\frac{e^{-x}}{\sqrt{x}}\right) , \qquad (B.20)$$

equation (B.19) becomes

$$-\frac{1}{U_o}\bar{u}_r\Big|_{r=a} = \frac{q}{p}\,\frac{[1 - \frac{3}{qa} - \frac{15}{128q^2a^2} + \cdots]}{[1 + \frac{1}{8qa} + \frac{9}{128q^2a^2} + \cdots]}$$

$$\therefore = \frac{q}{p}[1 - \frac{1}{2qa} - \frac{1}{8q^2a^2} + 0\,(\frac{1}{q^3a^3})] \quad . \tag{B.21}$$

Taking the inverse transform (Ab72), gives

$$-\frac{\partial u}{\partial r}\Big|_{r=a} = U_o\,[\frac{1}{\sqrt{\pi\kappa t}} - \frac{1}{2a} - \frac{1}{4a^2}\sqrt{\frac{\kappa t}{\pi}} + 0\,(\frac{\kappa t}{a^3})] \quad . \tag{B.22}$$

Hence,

$$\frac{I(t)}{A_o} = \frac{2\kappa}{a}\,[\frac{1}{\sqrt{\pi\,\kappa t}} - \frac{1}{2a} + 0\,(\frac{\sqrt{\kappa t}}{a^2})] \quad . \tag{B.23}$$

Compare this with (B.16); we see that for small values of $\kappa t/a^2 = T$, the following asymptotic approximation is valid:

$$\sum_{m=1}^{\infty} e^{-\xi_m^2 T} \sim \frac{1}{2\sqrt{\pi T}} \tag{B.24}$$

Similarly, from (B.15) we can evaluate the average concentration in the cylinder as a function of time:

$$v = \frac{2}{a^2}\int_0^a ru(r,t)dr = 4\,U_o\sum_{m=1}^{\infty}\frac{1}{\xi_m^2}\,e^{-\xi_m^2\frac{\kappa t}{a^2}} \quad . \tag{B.25}$$

But from (B.18) we also have

$$\bar{v} = \frac{2}{a^2}\int_0^a r\bar{u}dr = \frac{U_o}{p}[1 - \frac{2}{aq}\,\frac{I_1(qa)}{I_o(qa)}] \quad . \tag{B.26}$$

Using equations (B.19) and (B.20), this has the expansion

$$\bar{v} = \frac{U_o}{p} - \frac{2U_o}{apq}\left[1 - \frac{1}{2qa} - \frac{1}{8q^2a^2} + 0\left(\frac{1}{q^3a^3}\right)\right] . \qquad (B.27)$$

Taking the inverse transform, the average concentration is

$$v = U_o - U_o\sqrt{\frac{\kappa t}{a^2}}\left[\frac{4}{\sqrt{\pi}} + \sqrt{\frac{\kappa t}{a^2}} + 0\left(\frac{\kappa t}{a^2}\right)\right] . \qquad (B.28)$$

Compare this with (B.25); we see that for small values of $T = \kappa t/a^2$,

$$\sum_{m=1}^{\infty} \frac{1}{\xi_m^2} e^{-\xi_m^2 T} \sim \frac{1}{4} . \qquad (B.29)$$

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TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO. EPA 520/8-81-007	2.	3. RECIPIENT'S ACCESSION NO.
4. TITLE AND SUBTITLE Short- and Long-Term Leach Rates of Solidified Waste from a Cylindrical Container		5. REPORT DATE June 1981
		6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S) Kerry Landman		8. PERFORMING ORGANIZATION REPORT NO
9. PERFORMING ORGANIZATION NAME AND ADDRESS Office of Radiation Programs Environmental Protection Agency Washington, D.C. 20460		10. PROGRAM ELEMENT NO.
		11. CONTRACT/GRANT NO.
12. SPONSORING AGENCY NAME AND ADDRESS Same		13. TYPE OF REPORT AND PERIOD COVERED
		14. SPONSORING AGENCY CODE

15. SUPPLEMENTARY NOTES

16. ABSTRACT

The short- and long-term leach rates of radionuclides for the three-dimensional diffusive flow and for purely axial diffusive flow from a finite cylinder of solidified waste are determined here. These analytical results are compared with the ones obtained numerically by Hung for purely axial flow.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
radionuclides axial flow three-dimensional flow leach rates		

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS *(This Report)* Unclassified	21. NO. OF PAGES 26
	20. SECURITY CLASS *(This page)* Unclassified	22. PRICE

EPA Form 2220-1 (Rev. 4-77) PREVIOUS EDITION IS OBSOLETE