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Mass Transport in Salt Repositories:

Transient Diffusion into Interbeds

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The authors invite comments and would appreciate being notified of any errors in the report.

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1. Introduction

To estimate possible radioactive releases from a waste package to the near-field environment, we analyzed pressure-driven brine migration movement[1] and release rates of low-solubility and readily soluble nuclides by diffusion.[2] A possible pathway for radioactive release in salt repositories is interbeds and we have analyzed the steady-state transport of species through the interbeds in which there is ground-water flow.[3] A more realistic situation is when there is *no* ground-water flow in the interbeds. Here we use some results previously obtained for transient diffusion of radioactive species from a waste cylinder intersecting a planar fracture in rock[4] to the problem of diffusion from a waste cylinder intersecting an interbed in a salt repository.

2. Assumptions and Equations

The following assumptions are used.

- The crushed salt has consolidated around the waste cylinder.
- There is *no* ground-water flow in the interbed.
- The interbeds are planar and perpendicular to the longitudinal axis of the waste cylinder.
- The spacing between interbeds is constant.
- The waste cylinder is infinitely long. That is, end effects are ignored.
- Temperature effects are accounted for by using constant values of parameters such as diffusion coefficients, evaluated at the highest temperatures expected.
- Radionuclides can diffuse into the salt directly from the waste cylinder. Radionuclides can also diffuse into the interbed and then diffuse into the salt.

With these assumptions, the problem reduces to one of diffusion. Consider an infinitely long waste cylinder with radius $\hat{a}$ intersected by a planer interbed (Figure 1). We conservatively assume there is no metallic container and the surface concentration is the solubility limit of each species. The interbed width or thickness is $2\hat{b}$ and complete mixing across the interbed is assumed. Retardation is treated by equilibrium sorption. The mass balance for the time-dependent species concentrations in the salt and in the interbed are

$$\hat{K}_1\frac{\partial \hat{N}_1}{\partial \hat{t}} = \frac{\hat{D}_1}{\hat{r}}\frac{\partial}{\partial \hat{r}}\left(\frac{\partial \hat{N}_1}{\partial \hat{r}}\right) - \hat{\lambda}\hat{K}_1\hat{N}_1 - \frac{\hat{q}(\hat{r},\hat{t})}{\epsilon_1\hat{b}}, \qquad \hat{r} > \hat{a}, \hat{t} > 0 \tag{1}$$

$$\hat{K}_2\frac{\partial \hat{N}_2}{\partial \hat{t}} = \frac{\hat{D}_2}{\hat{r}}\frac{\partial}{\partial \hat{r}}\left(\hat{r}\frac{\partial \hat{N}_2}{\partial \hat{r}}\right) + \hat{D}_2\frac{\partial_2 \hat{N}_2}{\partial \hat{z}^2} - \hat{\lambda}\hat{K}_2\hat{N}_2, \qquad \hat{t} > 0, \hat{r} > \hat{a}, \hat{z} > 0 \tag{2}$$

in which

$$\hat{q}(\hat{r},\hat{t}) = -\epsilon_2\hat{D}_2\frac{\partial \hat{N}_2}{\partial \hat{z}}\bigg|_{\hat{z}=0}, \qquad \hat{t} > 0, \hat{r} > a \tag{3}$$

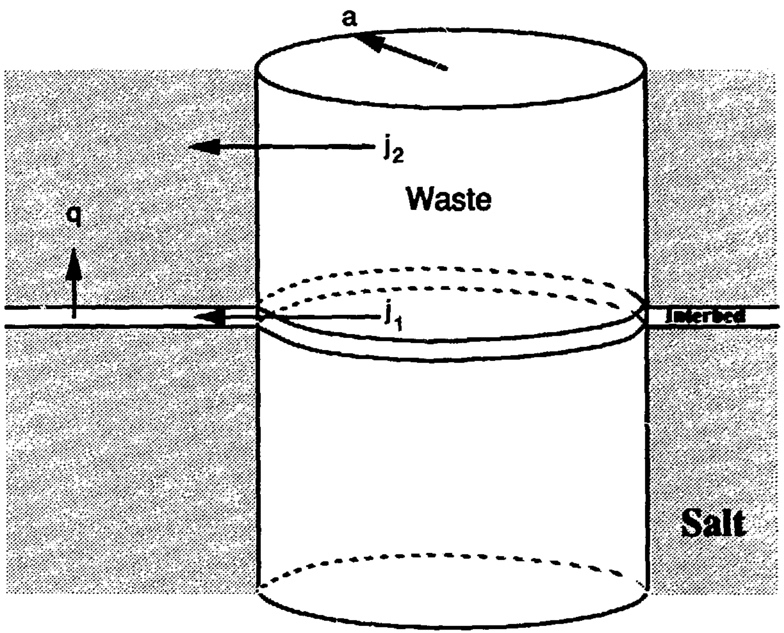

Figure 1. Waste Package Intersected by an Interbed

is the diffusive flux from the interbed to the salt through the interface [M/L^2-t]

where

subscript *1* denotes the interbed

subscript *2* denotes the salt

$\hat{N}_i$ is the liquid-phase species concentration [M/L^3]

$\hat{D}_i$ is the species diffusion coefficient [L^2/t]

$\hat{K}_i$ is the species retardation coefficient

$\hat{\lambda}$ is the decay constant [t^{-1}]

ϵ_i is porosity

$\hat{r}$ is the radial distance from the centerline of the waste cylinder [L]

$\hat{z}$ is the distance from the salt/interbed interface, in the direction normal to the interface [L]

$\hat{t}$ is time [t]

The side conditions are

$$\hat{N}_1(\hat{r},0) = 0, \qquad \hat{r} > \hat{a} \tag{4}$$

$$\hat{N}_2(\hat{r},\hat{z},0) = 0, \qquad \hat{r} > \hat{a}, \hat{z} > 0 \tag{5}$$

$$\hat{N}_1(\hat{a},\hat{t}) = \hat{N}^*, \qquad \hat{t} > 0 \tag{6}$$

where $\hat{N}^*$ is the solubility of the diffusing species

$$\hat{N}_1(\infty,\hat{t}) = 0, \qquad \hat{t} > 0 \tag{7}$$

$$\hat{N}_2(\hat{a},\hat{z},\hat{t}) = \hat{N}^*, \qquad \hat{z} > 0, \hat{t} > 0 \tag{8}$$

$$\hat{N}_2(\infty,\hat{z},\hat{t}) = 0, \qquad \hat{z} > 0, \hat{t} > 0 \tag{9}$$

$$\hat{N}_2(\hat{r},0,\hat{t}) = \hat{N}_1(\hat{r},\hat{t}), \qquad \hat{r} > a, \hat{t} > 0 \tag{10}$$

$$\left.\frac{\partial \hat{N}_2}{\partial \hat{z}}\right|_{\hat{z}\to\infty} = 0, \qquad \hat{r} > \hat{a}, \hat{t} > 0 \tag{11}$$

By introducing the following transformations

$$r \equiv \frac{\hat{r}}{\hat{a}} \tag{12}$$

$$z \equiv \frac{\hat{z}}{\hat{a}} \tag{13}$$

$$t \equiv \frac{\hat{D}\hat{t}}{\hat{K}_2\hat{a}^2} \tag{14}$$

$$\Delta \equiv \frac{\hat{D}_1\hat{K}_2}{\hat{D}_2\hat{K}_1} \tag{15}$$

$$b \equiv \frac{\epsilon_1\hat{b}\hat{K}_1}{\epsilon_2\hat{a}\hat{K}_2} \tag{16}$$

$$\lambda \equiv \frac{\hat{a}^2\hat{\lambda}\hat{K}_2}{\hat{D}_2} \tag{17}$$

$$N_1(r,t) \equiv \frac{\hat{N}_1(\hat{r},\hat{t})}{\hat{N}^*} \tag{18}$$

$$N_2(r,z,t) \equiv \frac{\hat{N}_2(\hat{r},\hat{z},\hat{t})}{\hat{N}^*} \tag{19}$$

$$q \equiv \frac{\hat{a}\hat{q}}{\epsilon_2\hat{D}_2\hat{N}^*} \tag{20}$$

where t is the Fourier number, eq. (1) and (2) can be made dimensionless as

$$\frac{\partial N_1}{\partial t} = \frac{\Delta}{r}\frac{\partial}{\partial r}(r\frac{\partial N_1}{\partial r}) - \lambda N_1 - \frac{q}{b}, \qquad r > 1, t > 0 \tag{21}$$

$$\frac{\partial N_2}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial N_2}{\partial r}\right) + \frac{\partial^2 N_2}{\partial z^2} - \lambda N_2, \qquad r > 1, z > 0, t > 0 \tag{22}$$

The full derivations and solutions are shown in Ahn *et al.*[4] (for the case of $\epsilon_1 = 1$). Only the analytic solution will be given here. The normalized diffusive flux from the waste cylinder to the interbed is

$$j_1(t) \equiv -\frac{\partial N_1}{\partial r}\bigg|_{r=1} = \sqrt{\lambda}\frac{K_1(\sqrt{\lambda})}{K_0(\sqrt{\lambda})} - \frac{2}{\pi}[I_1'(0,t) + I_2'(0,t) + I_4'(0,t)], \qquad t > 0 \tag{23}$$

And the normalized diffusive flux from the waste cylinder directly into the salt is

$$j_2(z,t) \equiv -\frac{\partial N_2}{\partial r}\bigg|_{r=1} = \sqrt{\lambda}\frac{K_1(\lambda)}{K_0(\sqrt{\lambda})} - \frac{2}{\pi}[I_1'(z,t) + I_2'(z,t) - I_3'(z,t) + I_4'(z,t)], \qquad z > 0, t > 0 \tag{24}$$

where

$$I_i'(z,t) = \int_0^\infty \bar{W}_i(s;z,t)\frac{s\,ds}{[M_0(s)]^2} \tag{25}$$

$$M_0(s) = \sqrt{[J_0(s)]^2 + [Y_0(s)]^2} \tag{26}$$

$$\bar{W}_1(s;z,t) = \frac{1}{\pi}\frac{(\Delta-1)\lambda}{\mu_2^2(\mu_1^2 - \frac{\mu_2}{b})}e^{z\mu_2}\mathrm{erfc}\left(\frac{z}{2\sqrt{t}} + \mu_2\sqrt{t}\right) \tag{27}$$

$$\bar{W}_2(s;z,t) = \frac{1}{\pi}\frac{(\Delta-1)\lambda}{\mu_2^2(\mu_1^2 + \frac{\mu_2}{b})}e^{-z\mu_2}\mathrm{erfc}\left(\frac{z}{2\sqrt{t}} - \mu_2\sqrt{t}\right) \tag{28}$$

$$\bar{W}_3(s;z,t) = \frac{2}{\pi}\frac{1}{\mu_2^2}e^{-\mu_2^2 t}\mathrm{erf}\left(\frac{z}{2\sqrt{t}}\right) \tag{29}$$

$$\bar{W}_4(s;z,t) = \frac{2}{\pi}\frac{\exp\left\{-\frac{z^2}{4t} - \mu_2^2 t\right\}}{\beta - \alpha}\left\{\frac{\beta\Delta - 1/b}{\beta^2 - \mu_2^2}\mathrm{H}\left(\beta\sqrt{t} + \frac{z}{2\sqrt{t}}\right) - \frac{\alpha\Delta - 1/b}{\alpha^2 - \mu_2^2}\mathrm{H}\left(\alpha\sqrt{t} + \frac{z}{2\sqrt{t}}\right)\right\} \tag{30}$$

$$\alpha = \frac{1-P}{2b} \tag{31}$$

$$\beta = \frac{1+P}{2b} \tag{32}$$

$$P = \sqrt{1 - 4b^2(\Delta - 1)s^2} \tag{33}$$

$$\mu_1^2 = s^2\Delta + \lambda \tag{34}$$

$$\mu_2^2 = s^2 + \lambda \tag{35}$$

$$\mathrm{H}(x) = e^{x^2}\mathrm{erfc}(x), \tag{36}$$

3. Numerical Illustrations

3.1 Input Data

We will now illustrate the above solution for a salt repository of nuclear waste. These parameter values are used. The waste cylinder is 0.31 m in radius.

Table I. Salt Properties

Parameter	Units	Salt	Interbed
Diffusion coefficient	cm^2/s	10^{-7}	10^{-7}
Porosity		0.001	0.01^5
Interbed half-width	m		0.01

Table II. Nuclide Properties

	Decay Constant (a^{-1})	Retardation Coefficient in Salt	Retardation Coefficient in Interbed
U-234	2.81×10^{-6}	20	1
Np-237	3.24×10^{-7}	20	1
Pu-239	2.84×10^{-5}	20	1

Some sensitivity analysis will be done.

In terms of the dimensionless quantities in eq. (13) to (17), we have

$$\Delta = \hat{D}_1\hat{K}_2/\hat{D}_2\hat{K}_1 = 20, \qquad b = 0.0161$$

3.2 Results: Flux into the interbed

We first calculate the mass flux into the interbed. Figures 2 and 3 show the dimensionless flux into the interbed as a function of the Thiele modulus, which is a dimensionless paramenter for radioactive decay, and at various Fourier numbers, which is dimensionless time. In Figure 2, at early times such as t=0.1 or about 600 years on the real-time scale, the fluxes into the interbed of all species are about the same, except for extremely short-lived ones. At larger t, such as t=10 or 100, long-lived species show markedly lower dimensionless fluxes. This is because for shorter-half-life species radioactive decay serves as an additional sink, increasing the gradient for dissolution.

Figure 3 shows the dimensionless flux into the interbed as a function of the Fourier number or dimensionless time for Pu-239 and U-234. These two long-lived species show identical mass fluxes up to t of about 20, then the shorter half-life of Pu-239 makes its flux slightly higher than that of U-234. The dimensionless fluxes of both species into the interbed reach steady state at about the time they diverge.

3.3 Results: Flux directly to the salt

Figures 4 to 6 show the dimensionless flux directly from the bare waste cylinder into the salt, as given by eq. (24). Figure 4 shows the dimensionless flux into the salt as a function of distance from the interbed/salt interface for different Fourier numbers. At t=0.1, the dimensionless flux in the vicinity of salt/interbed interface is smaller than that further away from the interface. This is due to the diffusion from the interbed to the rock matrix. Because the porosity in the interbed is higher than the porosity in the salt while the diffusion coefficient has been held constant, there is greater diffusive flux of the species in the interbed. The diffusion of the species from the interbed into the salt reduces the gradient for diffusion from the waste cylinder directly into salt, hence the lower flux closer to the salt/interbed interface. As time increases this region influenced by the interbed expands as shown in Figure 4.

Figure 5 shows the dimensionless flux as a function of the Fourier number for different Thiele moduli at a specific location, $z = 1$. At early time, all species show the same flux, but at later times, radioactive decay creates different gradients. The difference has been illustrated for three hypothetical species with Thiele moduli from 10^{-2} to 10^{-4}.

Figure 6 shows the effect of variation in interbed diffusion coefficient. For $\Delta = \hat{D}_1\hat{K}_2/\hat{D}_2\hat{K}_1$ and for a specific species with a fixed $\hat{K}_2/\hat{K}_1$, Δ is an indication of the effect of varying $\hat{D}_1$. As shown in Figure 6, the higher the interbed diffusion coefficient the higher the diffusion from the interbed into the salt, resulting in lower flux directly from the waste cylinder into the salt.

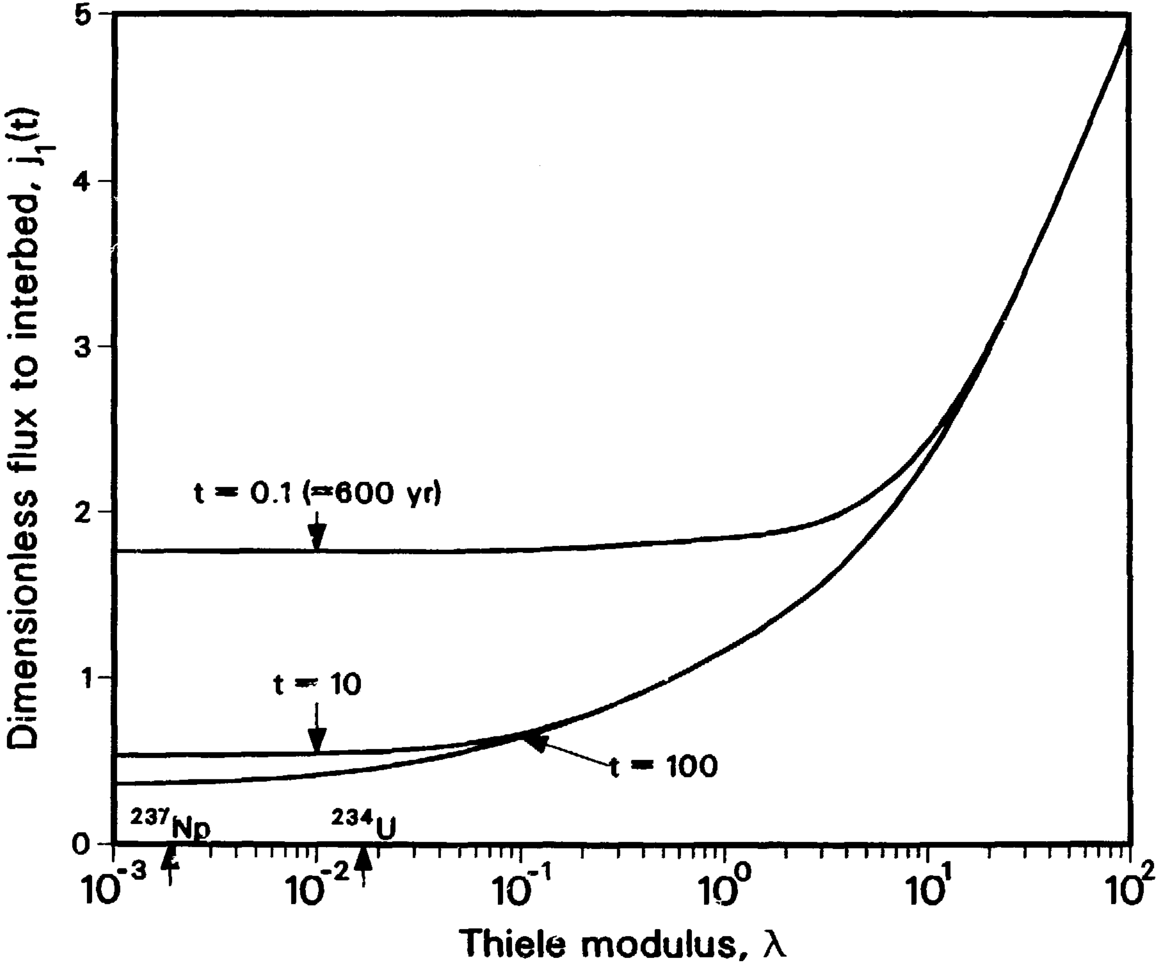

Figure 2. The Effect of Decay and Time on Mass Flux to the Interbed

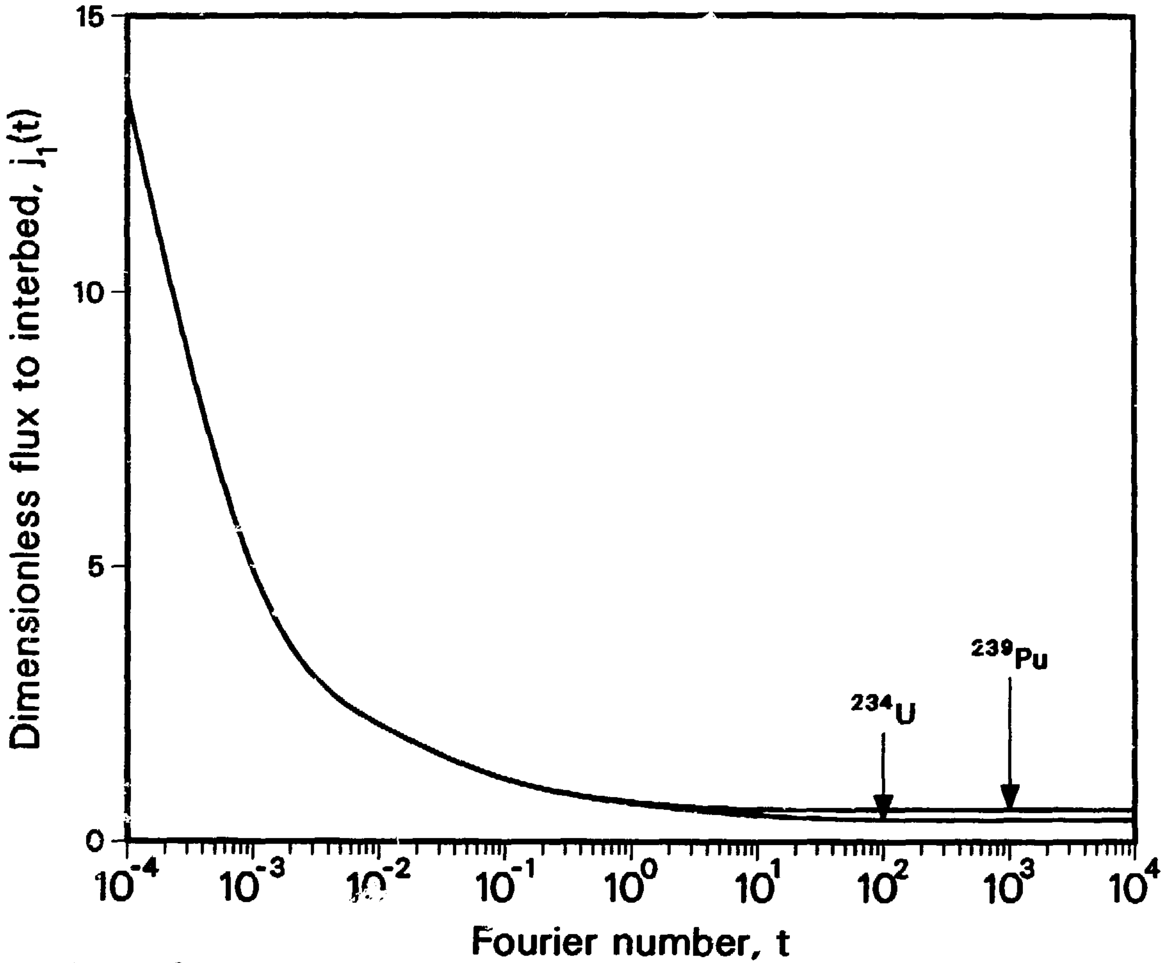

Figure 3. Dimensionless Flux of U-234 and Pu-239 to the Interbed

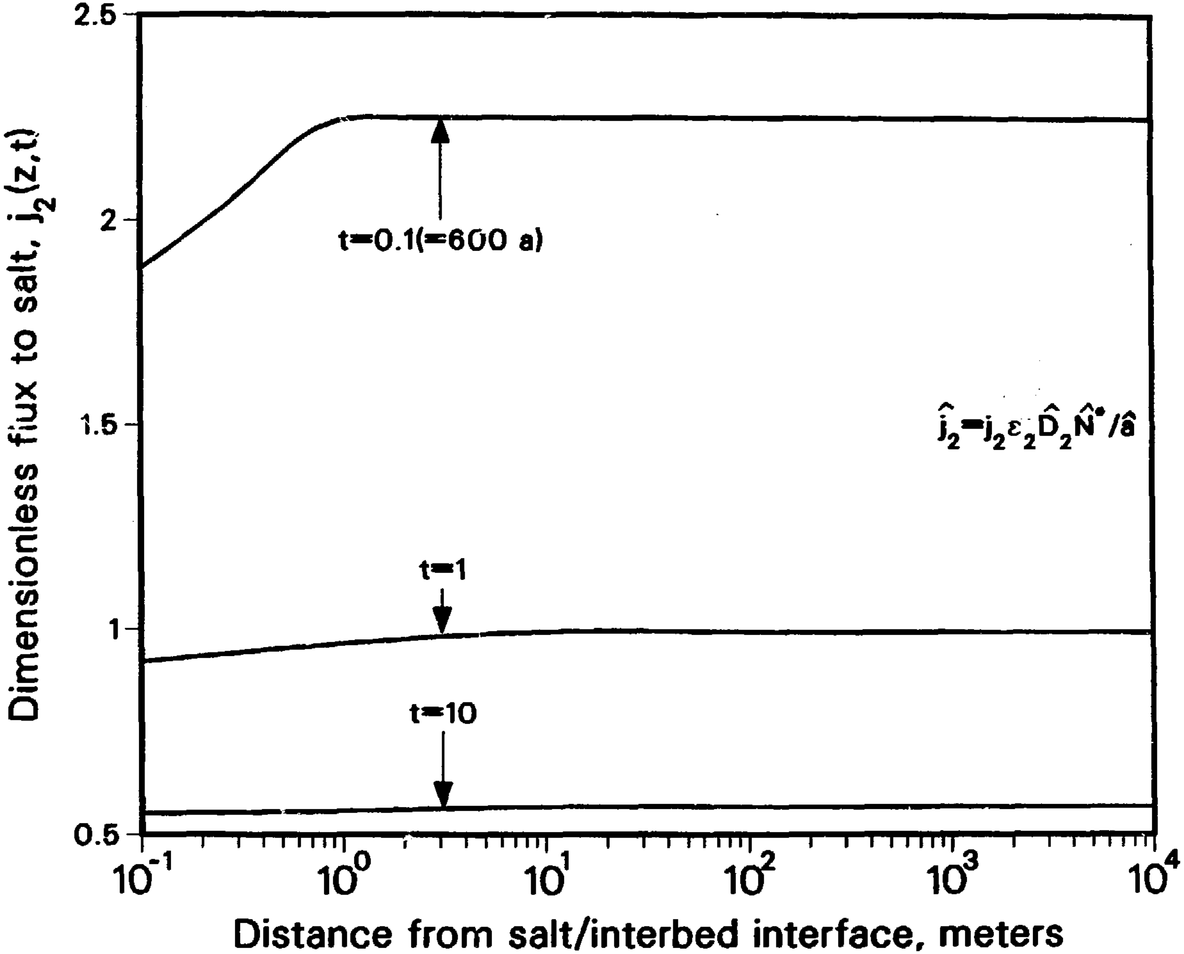

Figure 4. Diffusive Flux from the Waste Cylinder Directly into Salt

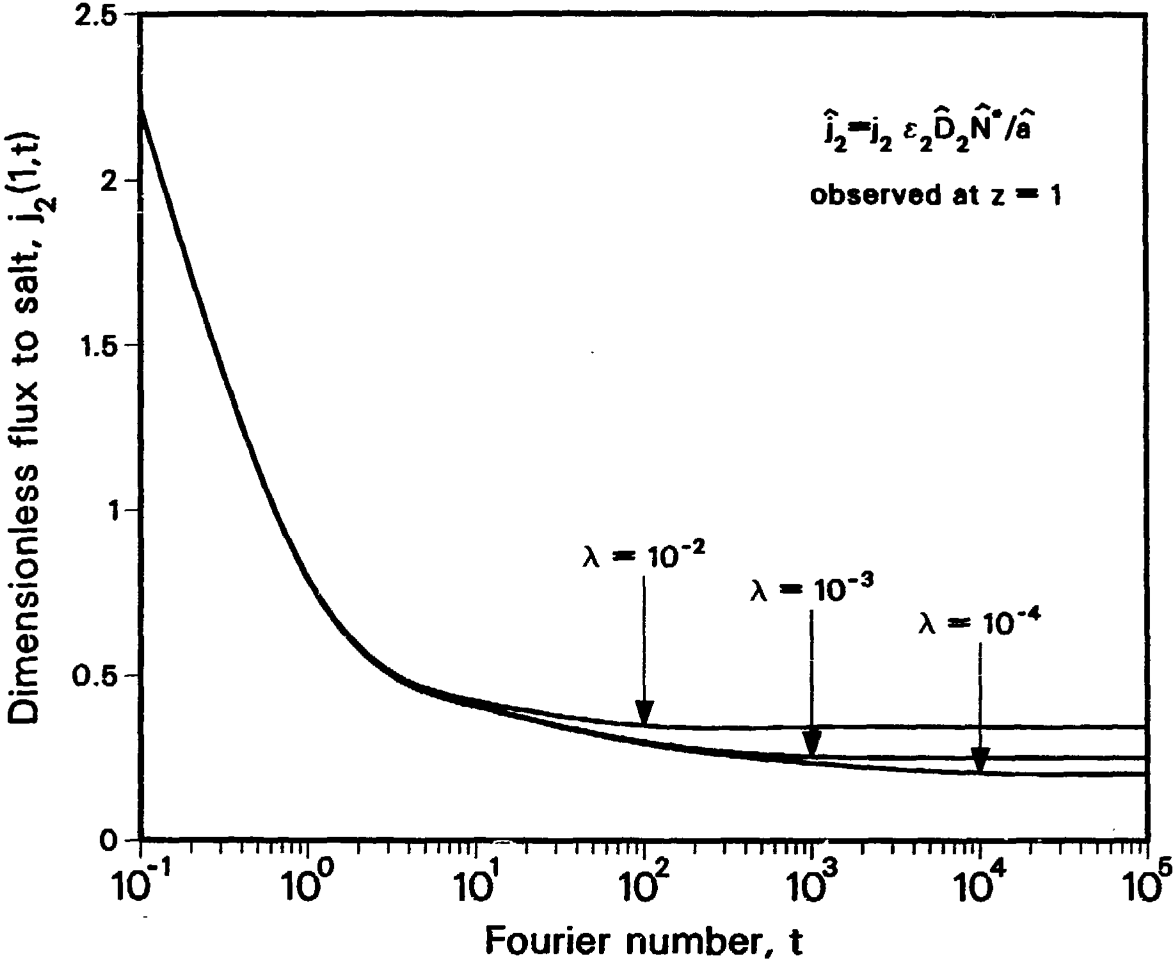

Figure 5. The Effect of Decay and Time on Mass Flux Directly to the Salt

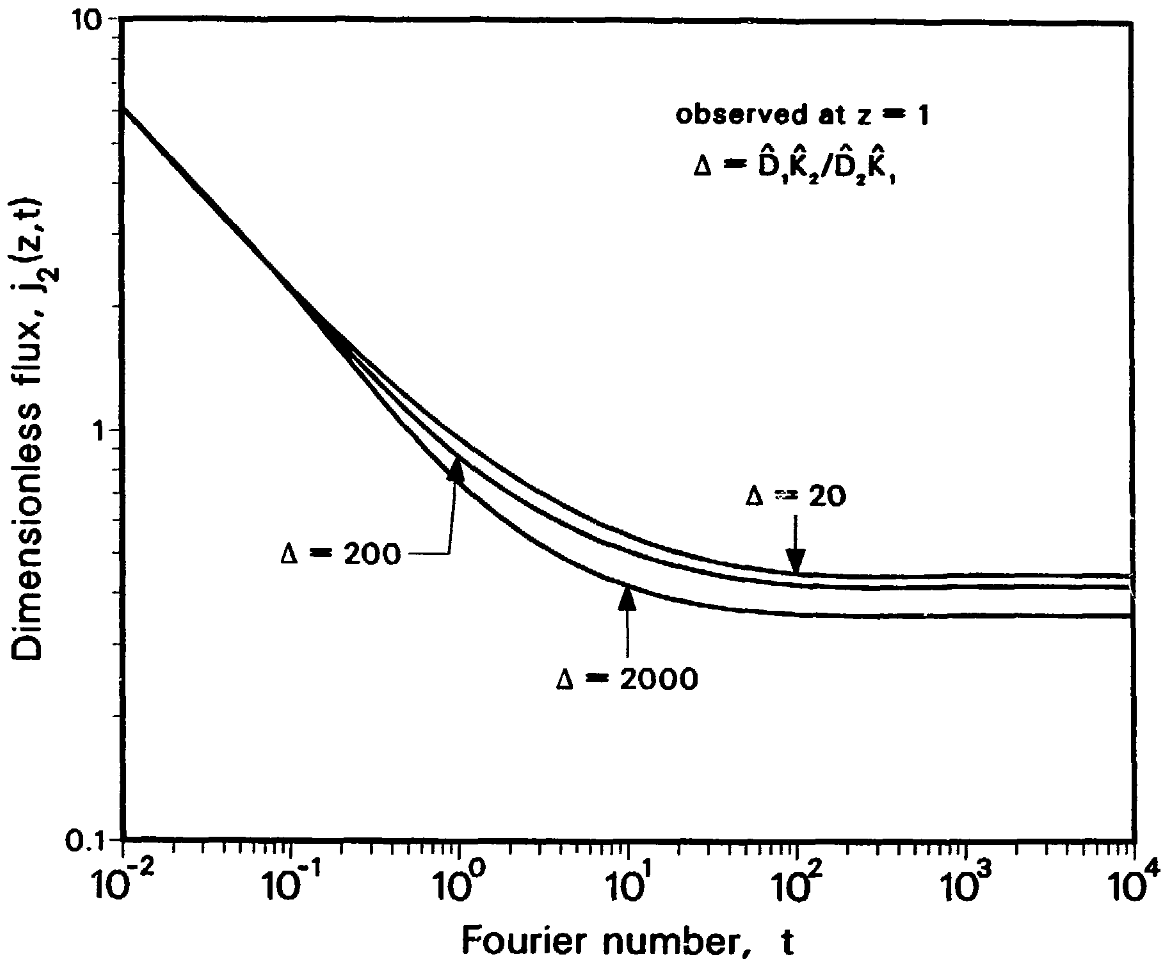

Figure 6. Dimensionless Flux Directly to Salt for Various Diffusion Coefficients

3.4 Results: Fractional release rate into the Interbed

The dimensional form of the mass flux, from eq. (23), can be expressed as

$$\hat{j}_1(\hat{a},\hat{t}) = \frac{\epsilon_1 \hat{D}_1 \hat{N}^*}{\hat{a}} j_1(t) \tag{37}$$

Then the total mass flux from a single waste package to an interbed of thickness $2\hat{b}$

$$\hat{J}_1(\hat{a},\hat{t}) = 4\pi\hat{a}\hat{b} \times \frac{\epsilon_1 \hat{D}_1 \hat{N}^*}{\hat{a}} j_1(t) \tag{38}$$

where the upper case $\hat{J}$ refers to a mass release rate with dimensions of mass per time. To calculate the fractional release rate of a species, we assume that the species k is released congruently with release of the matrix, and the matrix mass release rate can be calculated using eq. (38). Then, using the following equation of congruency

$$\frac{\hat{J}_{1,k}(\hat{a},\hat{t})}{\hat{J}_{1,m}(\hat{a},\hat{t})} = \frac{\hat{M}_k(\hat{t})}{\hat{M}_m(\hat{t})} \tag{39}$$

where

$\hat{J}_{1,m}(\hat{a},\hat{t})$ is the matrix mass release rate, [M/t]

$\hat{J}_{1,k}(\hat{a},\hat{t})$ is the species mass release rate, [M/t]

$\hat{M}_m(\hat{t})$ is the matrix inventory at time $\hat{t}$, [M]

$\hat{M}_k(\hat{t})$ is the species inventory at time $\hat{t}$, [M]

subscript k refers to the species

The fractional release rate of species k directly into the interbed, based on the species inventory at 1000 years, is

$$f_{1,k}(\hat{t}) = \frac{\hat{J}_{1,k}(\hat{t})}{\hat{M}_k(1000)} \tag{40}$$

The inventories are

$$\hat{M}_i(1000) = \hat{M}_i^o e^{-1000\hat{\lambda}_i} \qquad i = k, m \tag{41}$$

where

$\hat{M}_m^o$ is the initial inventory of the matrix [M]

$\hat{\lambda}_m$ is the decay constant of the matrix, [t^{-1}]

$\hat{M}_k^o$ is the initial inventory of the species [M] and

$\hat{\lambda}_k$ is the decay constant of the species [t^{-1}].

Substituting (39) in (40) we get

$$f_{1,k}(\hat{t}) = \frac{\hat{J}_{1,m}(\hat{a},\hat{t})}{\hat{M}_k^o e^{-1000\hat{\lambda}_k}} \frac{\hat{M}_k^o e^{-\hat{\lambda}_k \hat{t}}}{\hat{M}_m^o e^{-\hat{\lambda}_m \hat{t}}}$$

or

$$f_{1,k}(\hat{t}) = \frac{\hat{J}_{1,m}(\hat{a},\hat{t}) e^{-\hat{\lambda}_k(\hat{t}-1000)}}{\hat{M}_m^o e^{-\hat{\lambda}_m \hat{t}}} \tag{42}$$

In spent fuel we can neglect the decay of ^{238}U and set $e^{-\hat{\lambda}_m \hat{t}} = 1$.

The matrix inventory projected from the interbed is

$$\hat{M}_m^o = 2\pi \hat{a}^2 \hat{b} \hat{\rho} \tag{43}$$

where $\hat{\rho}$ is the matrix density [M/L^3]. For spent fuel, we use a value of 4.99×10^3 kg/m^3 for $\hat{\rho}$ and $\hat{N}^*$ of 10^{-3} g/m^3.

Eq. (42) now becomes

$$f_{1,k}(\hat{t}) = \frac{\hat{J}_{1,m}(\hat{a},\hat{t}) e^{-\hat{\lambda}_k(\hat{t}-1000)}}{2\pi \hat{a}^2 \hat{b} \hat{\rho}} \tag{45}$$

with the final result using (38)

$$f_{1,k}(\hat{t}) = \frac{2\epsilon_1 \hat{D}_1 \hat{N}^*}{\hat{a}^2 \hat{\rho}} e^{-\hat{\lambda}_k(\hat{t}-1000)} j_{1,m}(t) \tag{46}$$

The fractional release rate of ^{234}U into an interbed is shown in Figure 7. This result is for comparison with our previous steady-state result.[3]

3.5 Comparison with granite

Because this analysis was originally developed for transient diffusion from a waste cylinder into a rock fracture, there is some interest in comparing the overall releases from a waste cylinder facing an interbed in salt and a waste cylinder facing a fracture in granite. In this section, we compare the integrated releases from a waste cylinder of length ℓ.

The instantaneous mass flux into the interbed/fracture of thickness $2\hat{b}$ is, using (37),

$$\hat{m}_1(t) = 2\hat{b}\, 2\pi \hat{a} \hat{j}_1(\hat{a},\hat{t}) = 4\pi \hat{a} \hat{D}_2 \hat{N}^* \left(\frac{\epsilon_1 \hat{b} \hat{D}_1}{\hat{a} \hat{D}_2} \right) j_1(t) \tag{47}$$

and the instantaneous mass flux into the salt/rock is

$$\hat{m}_2(t) = 2 \times 2\pi \hat{a} \int_0^{\hat{\ell}/2-\hat{b}} \hat{j}_2(\hat{z},t) d\hat{z} = 4\pi \hat{a} \hat{D}_2 \hat{N}^* \epsilon_2 \int_0^{\hat{\ell}/2\hat{a}-\hat{\ell}/\hat{a}} j_2(z,t) dz \tag{48}$$

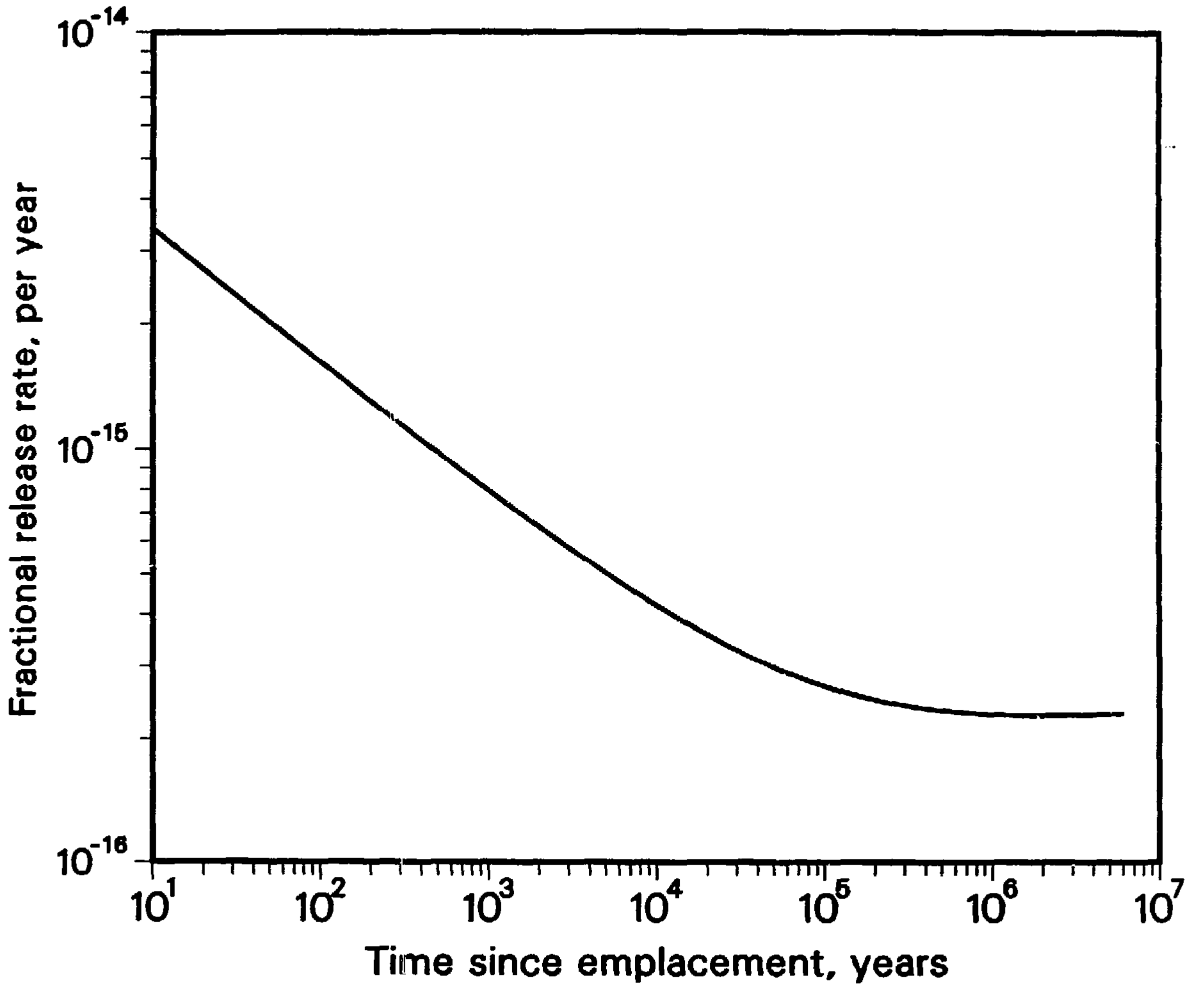

Figure 7. Fractional Release Rate of U-234 into an Interbed

Figure 8 shows the total mass transfer of a stable species, from a 3.65-m long waste cylinder into salt and granite using the following properties assumed for illustration.

Table III. Salt and Granite Data

	Porosity	Diffusion coefficient cm^2/s	Retardation coefficient	Solubility g/m^3
Salt	0.001	10^{-7}	20	10^{-3}
Salt Interbed	0.01	10^{-7}	1	10^{-3}
Granite	0.01	10^{-5}	500	10^{-3}
Granite Fracture	1	10^{-5}	1	10^{-3}

Because the product of porosity and diffusion coefficient of salt is approximately 10^{-3} times less than that of granite, the release rate to the surrounding salt is almost 10^{-3} times less than that of granite. Figure 8 shows that for a bare waste cylinder, the mass flux directly into the salt/rock is about 3 orders of magnitude higher than that into the interbed/fracture. However, it should be observed that a more realistic situation is for localized corrosion to expose waste only on contact with the interbed/fracture, and a partly degraded container will effectively cover the waste cylinder where it is in direct contact with the salt/rock.

4. Conclusions

We have calculated the dimensionless diffusive mass fluxes from an infinitely long bare waste cylinder in salt, facing an interbed. At the source a constant concentration boundary condition is imposed. If this concentration is the solubility, then this is a conservative analysis. We have also calculated fractional release rates into the interbed. All calculations show releases are low for the parameter values used in the numerical illustrations. The influence of radioactive decay has been demonstrated, as well as the interplay between diffusion from the waste cylinder directly into the salt and through the interbed into the salt.

We have also compared salt and granite as confining rocks for nuclear waste in the context of this analysis. Because the diffusion coefficient and porosity are lower in salt, the mass fluxes are also lower from a waste cylinder in salt.

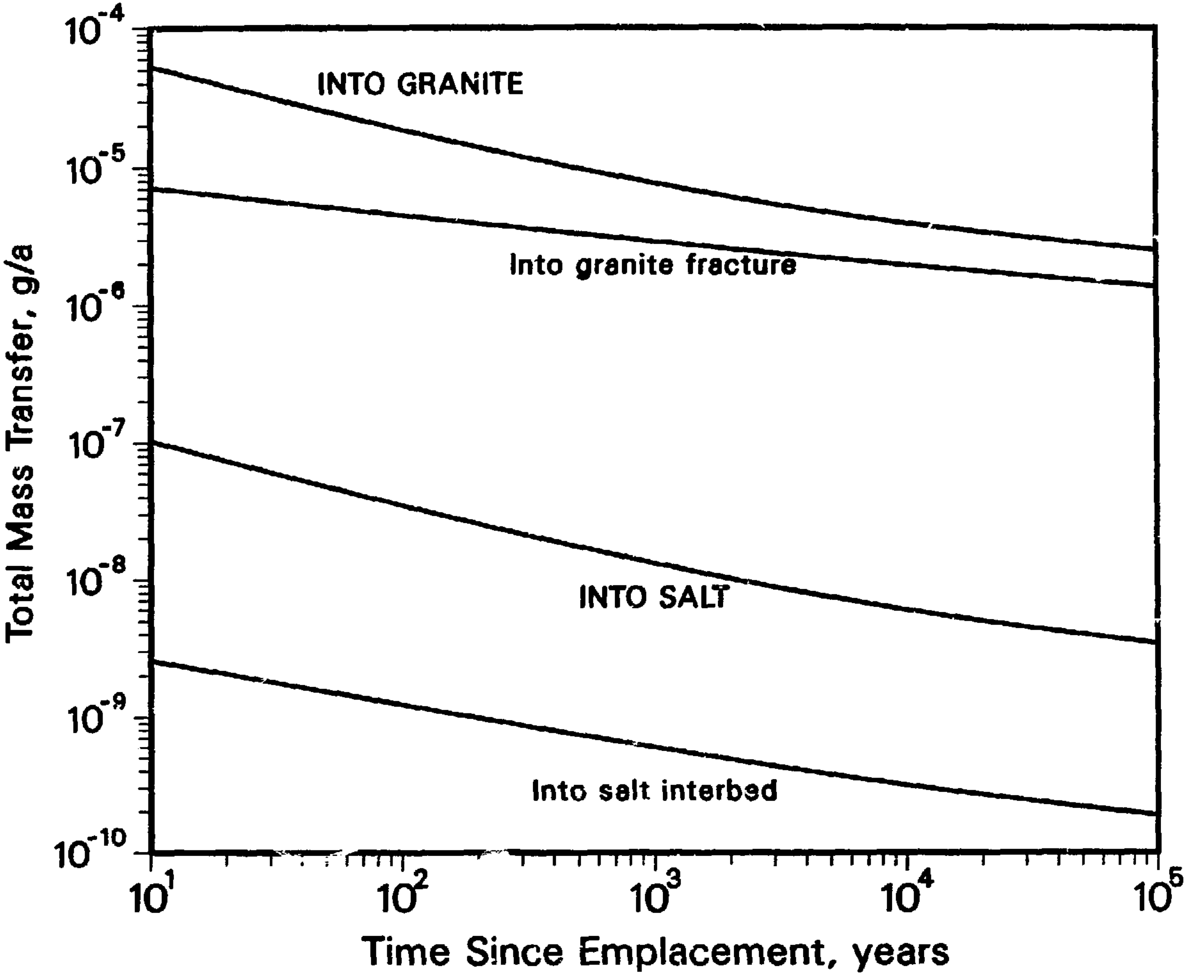

Figure 8. Total Mass Transfer of a Stable Nuclide from a 3.65-m Long Waste Cylinder in Salt and Granite

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