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Transient Diffusion From A Waste Solid Into Water-Saturated, Fractured Porous Rock¹

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Abstract

Numerical illustrations for transient mass transfer from an infinitely long cylinder intersected by a planar fracture are shown based on Chambré's exact analytical solutions. The concentration at the cylinder surface is maintained at the solubility. In the fracture contaminant diffuses in the radial direction. In the rock matrix three-dimensional diffusion is assumed in the cylindrical coordinate. No advection is assumed. Radioactive decay and sorption equilibrium are included. Radioactive decay enhances the mass transfer from the cylinder. Due to the presence of the fracture, the mass flux from the cylinder to the rock matrix becomes smaller, but the fracture effect is limited in the vicinity of the fracture in early times. Even though the fracture is assumed to be a faster diffusion path than the rock matrix, the larger waste surface exposed to the matrix and the greater assumed matrix sorption result in greater release rate to the matrix than to the fracture.

1 Introduction

This paper presents numerical results of an analytical study for mass transfer and transport of radionuclides released from a cylindrical waste solid into water-saturated, fractured porous rock. In this paper, we present, based on Chambré's exact analytical solutions, numerical results of the diffusive mass flux from a cylindrical waste solid into a planar fracture and into the surrounding rock matrix, for the low-flow conditions wherein near-field mass transfer is expected to be controlled by molecular diffusion [1].

Previous analytical studies [2,3,4,5,6] of the advective transport of dissolved contaminants through fractured rock have emphasized the effect of molecular diffusion in the rock matrix in affecting the space-time-dependent concentration of the contaminant as it moves along the fracture. Matrix diffusion only in the direction normal to the fracture surface was assumed (Figure 1(a)). Such studies illustrate the far-field transport features of fractured media. To predict the time-dependent mass transfer from a long

waste cylinder surrounded by porous rock and intersected by a fracture, the present study includes diffusion from the waste-solid surface directly into porous rock, as well as the more realistic geometry shown in Figure 1(b).

The problem was first proposed and solved analytically by Chambré. Analytical solutions have been derived for (1) the diffusive mass fluxes from the cylindrical waste solid into the fracture and into the rock matrix, (2) the diffusive mass flux across the rock/fracture interface, and (3) the concentrations of radionuclide in the fracture and in the rock matrix. For comparison of the cylinder model with the previous plane model, refer to [7].

2 Assumptions and Mathematical Formulation

We consider a cylindrical waste solid of infinite length and constant radius a [m], intersected by a planar fracture (Figure 1). To be conservative, we assume that no waste container is present. An infinitely long cylinder is a good approximation for a long cylinder with negligible end effects. A constant concentration N^* [kg/m³] of low-solubility dissolved species is pre-

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3 Numerical Evaluations

We assume that the waste solid comes from the spent fuel of a pressurized water reactor. The radius of the cylinder is $\hat{a} = 25$ cm. Fracture width $2\hat{b}$ is 1 cm. Surrounding rock has porosity $\varepsilon_2 = 0.01$, whereas the fracture has no filling material ($\varepsilon_1 = 1$). The diffusion coefficient is the same for both fracture and rock, and is conservatively chosen as that for a liquid continuum ($500 \text{ cm}^2/\text{yr}$). Sorption on fracture walls is neglected ($\hat{K}_1 = 1$) while sorption in the rock matrix is assumed to retard the matrix diffusion process by the factor $\hat{K}_2 = 500$. Three actinides and a stable nuclide are compared in the numerical results: $\lambda = 2.806 \times 10^{-6} \text{ yr}^{-1}$ for ^{234}U , $\lambda = 1.513 \times 10^{-3} \text{ yr}^{-1}$ for ^{241}Am , and $\lambda = 2.841 \times 10^{-8} \text{ yr}^{-1}$ for ^{239}Pu .

With these values, non-dimensionalized parameters can be calculated as: $\hat{b} = 0.004$, $\Delta = 500$, $\hat{t} = 1.6 \times 10^{-3} \hat{t}$ where \hat{t} is measured in the unit of years, and $\lambda = 1.754 \times 10^{-3}$ for ^{234}U , $\lambda = 0.9456$ for ^{241}Am , and $\lambda = 1.7755 \times 10^{-2}$ for ^{239}Pu .

Figure 2 gives an overall idea of how the contaminant is transferred from the cylinder and is transported in the fractured porous rock. There shown are instantaneous concentration isopleths, mass flux from the cylinder to rock, mass flux to fracture, and flux across the rock/fracture interface for ^{239}Pu at Fourier number $\hat{t} = 1$, which corresponds to 625 yr for the parameter values shown above. The concentration in rock is larger near the cylinder and near the fracture. The concentration isopleth shows that the influence of the fracture on the concentration in the rock matrix becomes negligible beyond five radii from the rock/fracture interface. The mass flux from the cylinder into the fracture is calculated to be about two orders of magnitude greater than that into the rock matrix because of the assumed hundred-fold greater porosity in the fracture. The mass flux into the rock matrix becomes smaller in the vicinity of the fracture because contaminant diffusing from the fracture reduces the concentration difference between the surface of the cylinder and inside the rock matrix.

The mass flux across the interface between rock and fracture is zero at the surface of the cylinder because of the boundary conditions at $r = 1$. The concentration difference increases with the distance from the cylinder surface because of the assumed larger retardation factor and smaller porosity in rock matrix, so concentration in the rock matrix decreases faster with distance than in the fracture. Mass flux across the interface starts to decrease after it reaches the maximum because both concentrations in the fracture and in the rock matrix approach zero. Detailed numerical investigation indicates that the maximum

value decreases and that the location of the maximum advances with time. Because the both concentrations approach to the steady state, the profile for $\hat{t} = 10^4$ shows slight change from $\hat{t} = 10^2$.

Figure 3 shows the changes of mass flux from the cylinder to the fracture with Fourier number for three actinides and a stable nuclide. In very early times effect of decay is not apparent. Curves for shorter-half-life nuclides deviate from that for a stable nuclide at an earlier time and reach steady state. For a stable nuclide the mass flux approaches to zero as time increases. Because of the loss by radioactive decay during diffusion in the medium, less will reach given distance \hat{r} or the surface for shorter half-life nuclides. There are in the steady state the concentration gradients becomes steeper for shorter half-life nuclides. Thus, radioactive decay enhances the mass transfer from the cylinder to the rock matrix and to the fracture.

In Figure 4 the amount of radionuclide released into rock matrix is compared with that into the fracture. Release rates are normalized by the factor $4\pi\hat{a}\hat{D}_2\hat{N}^0$. Even though the mass flux from the waste into the rock matrix is low relative to that into fracture, the larger waste surface exposed to the matrix and the greater assumed matrix sorption result in greater release rate to the matrix than to the fracture. This indicates that for the parameters assumed here, the earlier mass-transfer theory [4] for a waste solid completely surrounded by porous rock can adequately predict release rates in low-flow conditions in fractured rock. If tortuosity significantly reduces the diffusion coefficient in the rock matrix and not in the fracture, mass-transfer directly from the waste to the fracture becomes more important.

4 Conclusions

Mass fluxes to the fracture and to the rock matrix reach steady state for radioactive contaminants. Shorter half-life nuclides reach steady state earlier. In very early times the effect of radioactive decay is negligible. Radioactive decay enhances mass transfer from the cylindrical waste solid, from the comparison of the magnitudes of the steady-state mass fluxes.

Because of diffusion from fracture to rock, the mass flux from the cylinder to rock matrix is smaller in the region near the fracture than in regions far from the fracture. The effect of the presence of the fracture is limited to a shallow region in the rock matrix in early times. From the numerical result for ^{239}Pu , after a Fourier number of 100 the fracture effect on the mass flux from the cylinder to rock matrix can

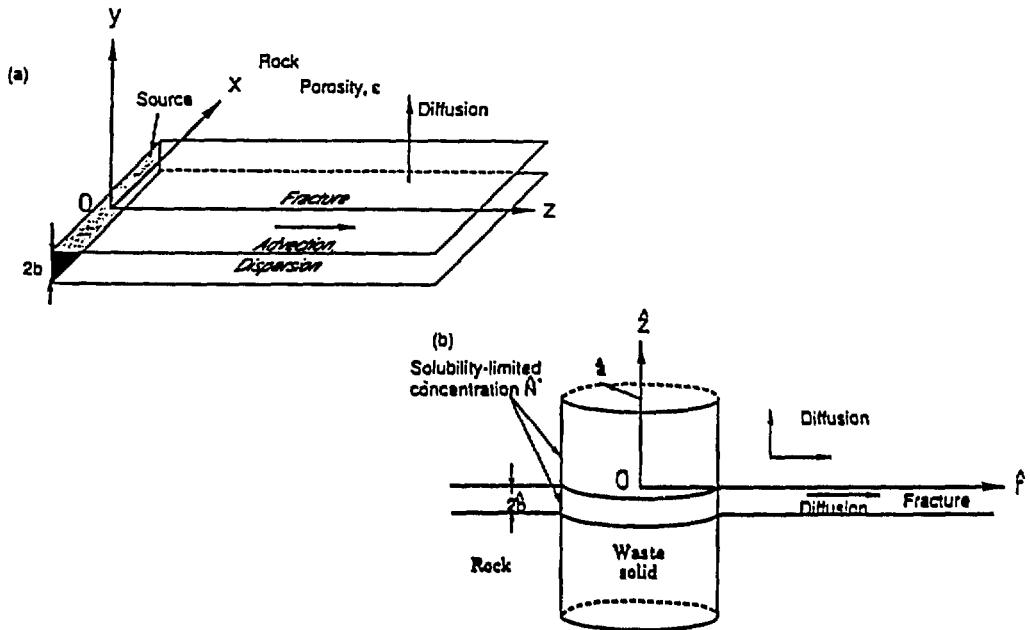


Figure 1: Geometry and physical processes considered in the model with a single, planar, infinite fracture (a) and cylindrical geometry with three-dimensional diffusion in fracture (b). Angular symmetry is assumed for (b).

scribed in the water at the waste surface. The contaminant undergoes molecular diffusion both in the fracture and in the rock matrix. The porosities of fracture and rock are ϵ_1 and ϵ_2 . Pores are saturated with stationary water. Notice that we assume that in some cases the fracture is filled with porous sediment. If ϵ_1 is unity, then the fracture is completely open. Width of the fracture, $2b$ [m], is considered to be much smaller than other dimensions such as the cylinder radius, so we assume complete mixing across the width. Then, the concentrations in the fracture and in the rock matrix are described in two-, and three-dimensional cylindrical coordinate systems, respectively. Different properties are assumed for the local sorption equilibrium in the fracture and in the rock matrix. Radioactive decay is included without any precursors. We assume angular symmetry. To simplify mathematical manipulations, we introduced the following non-dimensionalization:

$$t \equiv \frac{\hat{D}_2 i}{\hat{K}_2 \hat{a}^2}, \quad (1)$$

$$\Delta \equiv \frac{\hat{D}_1 \hat{K}_2}{\hat{D}_2 \hat{K}_1}, \quad \hat{b} \equiv \frac{\hat{a} \hat{K}_1}{\epsilon_2 \hat{a} \hat{K}_2}, \quad \lambda \equiv \frac{\hat{a}^2 \hat{\lambda} \hat{K}_2}{\hat{D}_2}$$

where \hat{D}_i is diffusion coefficient of the nuclide in region i [m^2/yr], $\hat{\lambda}$, radioactive decay constant [yr^{-1}], ϵ_i , porosity of region i , dimensionless, i , time [yr], and \hat{K}_i , the retardation factor for region i , dimensionless, $i = 1, 2$. Subscripts 1 and 2 refer to fracture

and rock, respectively. The hat symbol indicates the quantities with physical dimensions. The variable t is the Fourier number, which measures the time of the diffusion process. λ is the Thiele modulus. Concentrations in the fracture and in the rock matrix are normalized by the solubility, \hat{N}^* .

The governing equations and the side conditions for the space-time-dependent concentrations in the fracture and in the rock matrix for the non dimensionalized system can be written by following the above description on the physical processes considered. Chambré solved the problem analytically. He also derived the expressions for the dimensionless diffusive fluxes of the nuclide at the waste surface from the waste to the fracture, $\epsilon_1 j_1(t)$, and to the porous rock, $\epsilon_2 j_2(z, t)$. The analytical solutions are quite lengthy, and are not shown here. For more details about the solution method and the analytical solutions, refer to [8].

We have three cases depending on the value of Δ : $\Delta > 1$, $\Delta = 1$, and $\Delta < 1$. By definition of Δ , (2), $\Delta > 1$ means that diffusion is faster in the fracture than in the rock matrix, which can be considered as the most likely case. The case $\Delta < 1$ could happen if the fracture is filled with highly sorbing material such as clay. In the case of $\Delta = 1$, the waste cylinder is surrounded entirely by homogeneous porous rock. The solution to $N_1(r, t)$ becomes identical to that to $N_2(r, z, t)$. From the standpoint of safety assessment of waste disposal, $\Delta > 1$ can be considered as the most undesirable case. So we assume $\Delta > 1$ below.

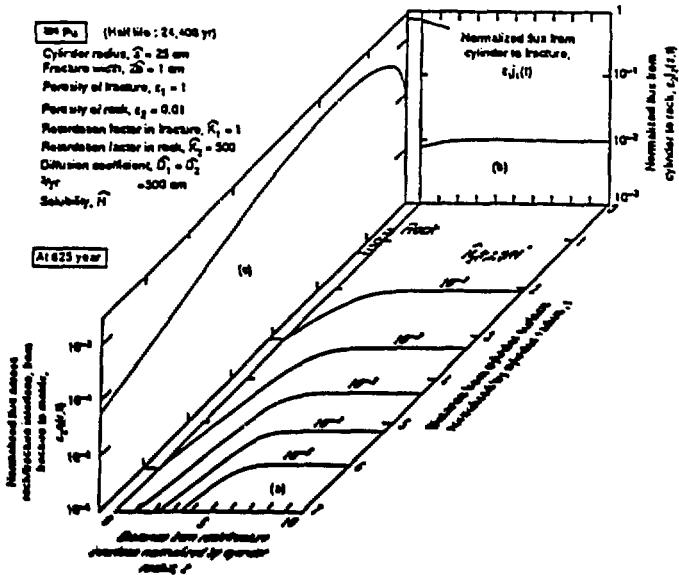


Figure 2: Illustration of migration of ^{239}Pu at 625 year, or Fourier number 1 (a) isopleths of normalized concentrations in fracture and in rock, (b) normalized flux from waste cylinder to rock, and (c) normalized flux from fracture to rock.

be neglected, and the mass flux to the rock matrix becomes virtually constant over the waste surface.

The present model is applicable until the nuclide, which initially exists in the slice of the cylinder exposed to the fracture, is all released into the fracture.

Although the mass flux to the fracture can be as much as hundred-fold larger than that to the rock matrix because of complete openness of the fracture, the total amount of the nuclide released into the rock matrix at a certain time becomes in some cases greater than that to the fracture because the waste surface in contact with the rock matrix is much larger than that in contact with the fracture. Under such conditions the model for the cylinder surrounded completely by rock matrix is adequate.

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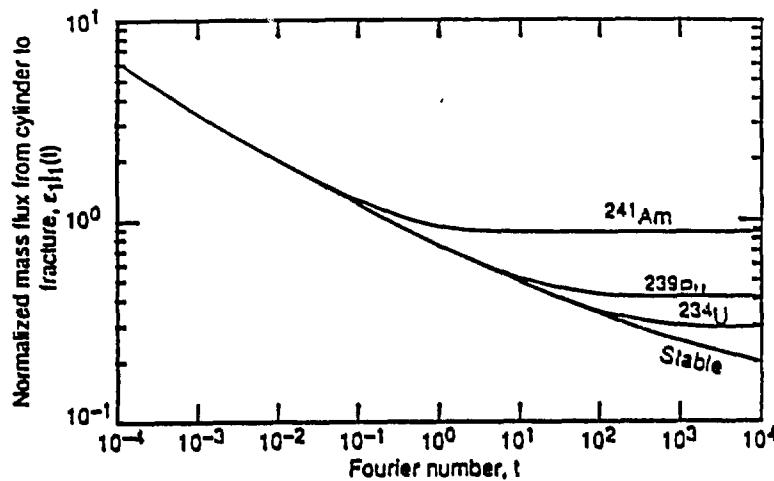


Figure 3: Mass fluxes for three radionuclides and a stable nuclide from the cylinder to the fracture at the cylinder surface as a function of time. Parameters from Figure 2 apply.

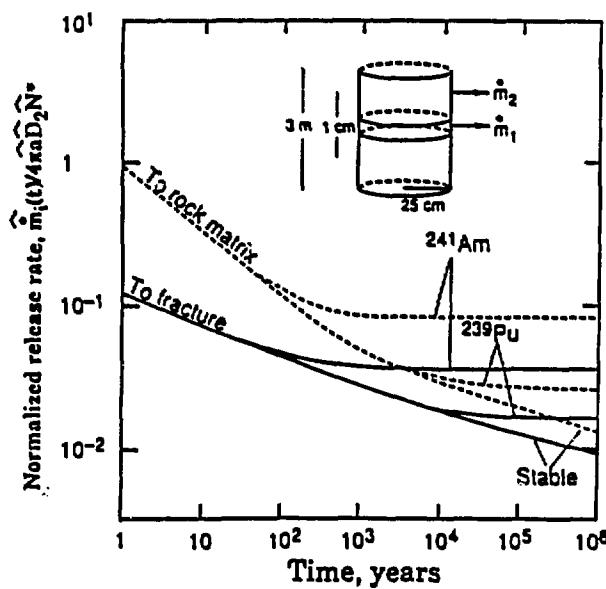


Figure 4: Normalized release rates of nuclides from a 3m-high waste cylinder. Geologic parameters from Figure 2 apply.

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