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Effects of Limiting Matrix Diffusion of Radionuclide in Fractured Porous Rock: Numerical Inversion of Laplace Transform

Jin Beak Park* and Kun Jai Lee
Korea Advanced Institute of Science and Technology
373-1 Kusong-dong, Yusong-gu
Taejon 305-701 Korea

Youngsoo Hwang
Korea Atomic Energy Research Institute
150 Dekjin-Dong, Yousong-Gu
Taejon 305-353 Korea

Abstract

To identify the effect of the limiting zone for the matrix diffusion from an open fracture into surrounding porous medium recently found from field studies, numerical solutions are evaluated. The numerical inverse solutions are then verified with existing solution of infinite diffusion into porous rock. Numerical results with band release mode illustrate that for smaller thickness of permeable zone, the effect of a limiting capacity of matrix diffusion by the existence of an impermeable boundary becomes noticeable.

1. Introduction

In the assessment for the long-term performance of deep geologic repository in crystalline rocks such as granite and tuff, fractures have been considered as one of the prime pathways of groundwater flow and consequent radionuclide transport.

In 1970's, transport in a fracture network was idealized as a single fracture without matrix diffusion so that it is identical to the flow in a pipe [1-2]. Then the matrix diffusion contributing to the delay of radionuclide transport to the infinite extend of porous rock matrix [3-9] was included in the model.

To represent the discrete fractures, more robust numerical models [10-17] were developed especially through the international Stripa Project [18] and the following Aspo Project [19]. In these model development exact mechanism of matrix diffusion was not fully considered to avoid the mathematical complexity. Instead, these models focus on the effect of fracture networks where fractures intersect fractures to create pathways for groundwater flow and radionuclide transport.

Recent field studies [20-22], however, have challenged the traditional concept of the matrix diffusion into the surrounding rock. Unlikely the previous concept of the diffusion into the infinitely long porous medium, field survey illustrates that only a small portion of the surrounding rock adjacent to a fracture is applicable for the matrix diffusion and the rest of the rock is, in practice, impermeable against the diffusion as shown in Fig. 1. In current model, the retardation mechanism on the radionuclide transport in a fracture by matrix diffusion is overestimated, because only a smaller portion rock contributes to "*suck and hold*" the radionuclides.

Therefore, it is worthwhile to estimate what would be the real impact of the "*limiting matrix diffusion*" concept by new mathematical modeling. Conclusions from a new study shall be either: (1) To confirm that this newly

* Nuclear Environmental Technology (KEPCO)

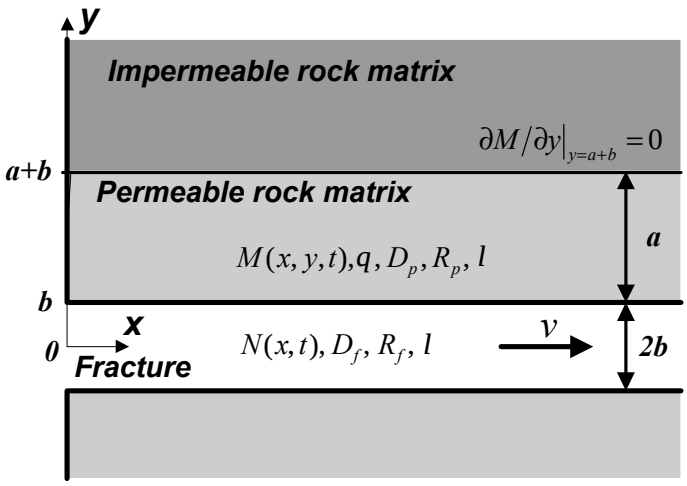

Figure 1 Physical system for the limiting matrix diffusion from a single, planar fracture into a finite rock matrix with a thickness, *a*

found mechanism is so important that it should be either included in the overall performance assessment of a potential repository or models without matrix diffusion should be applied for conservative assessment, or (2) To identify that the effect of the limiting matrix diffusion can be safely ignored.

In this study, the effect of limiting matrix diffusion from a fracture into a porous rock was assessed by numerical inversion of the Laplace Transform. Normalized concentrations of radionuclide in a fracture and a surrounding matrix was calculated and compared with those with infinite matrix diffusion.

2. Physical System

The real physical system has been idealized as shown in Fig. 1. The fracture is idealized as a planar one with an aperture of *2b*. The surrounding rock is simplified as a fully saturated porous medium with an effective porosity of *q*, [-].

The transport mechanisms in a fracture are (1) advective transport along the fracture, (2) longitudinal hydrodynamic dispersion in the direction of the fracture axis, and (3) molecular diffusion into the rock matrix. Inside the rock, since the permeability is so low diffusion is the only prime transport mechanism.

To avoid mathematical complexity, longitudinal diffusion in the rock matrix is assumed not to contribute to the migration of contaminants in a porous medium. Since a fracture is fully filled with groundwater, no sorption is considered in a fracture expect for the one onto the interface of a fracture and a surrounding medium. Inside a rock, rather strong sorption occurs. In this modeling no decay chain is considered.

To make a problem mathematically simple, some additional assumptions with certain rationales are made. First, the constant advective velocity is assumed from the fully developed flow theory [23]. Even though advective transport is considered in a fracture due to the characteristic of a potential repository rock, the flow velocity is still limited to laminar so that the more complex turbulent concept can be avoided and the traditional Darcy' s law can be applied.

The fracture aperture is much smaller than the fracture length so that after a certain distance from a fracture inlet, the flow is fully developed. In that sense, it is reasonable to take the pore water velocity as constant. Also, if an aperture is so thin, the flow is assumed to be well mixed across a fracture so that the *y*-dependency inside a fracture can be ignored in mathematical modeling. From a geo-chemical point of view, an instant local chemical equilibrium is assumed between water and solid phase.

3. Mathematical Formulation

From the mass balance theory with the above assumptions, the governing equation for transport of a nuclide in a fracture is derived [9] as:

$$R_f \frac{\partial N}{\partial t} - D_f \frac{\partial^2 N}{\partial x^2} + v \frac{\partial N}{\partial x} + R_f l N + \frac{q}{b} = 0, x > 0, t > 0, \quad (1)$$

where,

$N(x,t)$ = concentration of the radionuclide in fracture water [kg-nuclide/m3-water],

R_f = retardation coefficient on a fracture-matrix interface, [-],

D_f = longitudinal dispersion coefficient [m2/yr],

l = decay constant [1/yr],

$q(x,t)$ = diffusive flux of a contaminant from a fracture into a rock matrix [kg-nuclidee/m2 yr] defined as:

$$q = -q D_p \partial M / \partial y \big|_{y=b}, x > 0, t > 0, \quad (2)$$

where,

$M(x,y,t)$ = concentration of the radionuclide in rock matrix water, [*kg-nuclide/m^3-water*],

D_p = pore diffusion coefficient in a rock matrix, [*m^2/yr*].

Similarly, the governing equation describing the movement of a radionuclide in the rock matrix is:

$$R_p \frac{\partial M}{\partial t} - D_p \frac{\partial^2 M}{\partial y^2} + R_p l M = 0, x > 0, b < |y| < a + b, t > 0, \quad (3)$$

where,

R_p = retardation coefficient in a rock matrix, [-].

Side conditions of Eqs. (1) and (3) are as:

$$N(x,0) = 0, x > 0, \quad (4)$$

$$M(x,y,0) = 0, x > 0, b < |y| < a + b, \quad (5)$$

$$N(0,t) = N_o e^{-lt}, t > 0, \quad (6)$$

$$N(\infty,t) = 0, t > 0, \quad (7)$$

$$M(x,b,t) = N(x,t), x > 0, t > 0, \quad (8)$$

$$\partial M(x,y,t)/\partial y \big|_{y=a+b} = 0, x > 0, t > 0. \quad (9)$$

where N_o is the source concentration at the fracture inlet.

Eqs. (4) and (5) indicate zero initial conditions. Eq. (6) describes the decay inlet boundary condition and Eq. (7) depicts the zero boundary condition at the outlet. Eq. (8) explains the concentration continuity at the fracture and rock interface. Eq. (9) is the key of the problem. It stipulates the limiting capacitance of the matrix diffusion at "*a*" meters into the rock from the fracture and matrix interface. Beyond this point, the rock is practically "*impermeable*" so that no nuclides can penetrate into any further.

Applying the Laplace transform of Eq. (3) and solve ordinary differential equation (O.D.E) with the Laplace-transformed side conditions, the Laplace solution of Eq. (3) can be written as:

$$\tilde{M} = \tilde{N} \frac{\cosh(B\sqrt{p + l}\,(y - a - b))}{\cosh(aB\sqrt{p + l})}, x > 0, b \le |y| \le a + b, \quad (10)$$

$$B = \sqrt{R_p / D_p}\,. \quad (11)$$

The tilde symbol, ~, stands for the Laplace-transformed variable and p is the Laplace variable in a complex domain.

Applying the Laplace transform to Eq. (1) with the initial condition in Eq. (4) and substituting Laplace-

transformed diffusive flux into the resultant equation yields:

$$\frac{d^2\bar{N}}{dx^2}-\frac{v}{D_f}\frac{d\bar{N}}{dx}-\frac{R_f}{D_f}[(p+l)+a\sqrt{p+l}\tanh(aB\sqrt{p+l})]\bar{N}=0, x>0, D_f\neq 0, \quad (12)$$

$$a=q\sqrt{R_pD_p}/bR_f. \quad (13)$$

Eq. (12) can be solved with the following Laplace transformed boundary conditions:

$$\bar{N}(0,p)=\frac{N_o}{p+l}, \quad (14)$$

$$\bar{N}(\infty,p)=0. \quad (15)$$

The final Laplace-transformed normalized solution for *N(x,t)* is depicted as:

$$\frac{\bar{N}(x,p)}{N_o}=\frac{1}{p+l}e^{bx}\exp[-bx\sqrt{1+Y(p+l)+Ya\sqrt{p+l}\tanh(aB\sqrt{p+l})}], x\geq 0, D_f\neq 0, \quad (16)$$

$$b=v/2D_f \text{ and } Y=4D_fR_f/v^2. \quad (17) \text{ and } (18)$$

Eqs. (10) and (16) can be directly used to estimate the radionuclide concentrations at certain distances and at certain times. This can be achieved by the numerical inversion of the Laplace transform.

If a band release mode with leaching time, *T*, is considered, Eq. (6) is rewritten as:

$$N(0,t)=N_oe^{-lt}[h(t)-h(t-T)], t>0. \quad (19)$$

The superposition technique can be used to find the general solution, which is subject to the inversion of Eq. (16), *N(x,t)*:

$$N^b(x,t)=N(x,t)-e^{-lT}N(x,t-T)h(t-T), \quad (20)$$

where superscript *b* indicates the band release solution and *h(t)* means the Heavyside step function, [-].

4. Results and Discussions

Input parameters for the numerical evaluation are summarized in Table 1. Numerical inversion technique suggested by Talbot[24] is used to obtain normalized radionuclide concentration in fracture, $N(x,t)/N_o$.

Figs. 2 and 3 illustrate the normalized concentrations as a function of the distance from a fracture inlet at fixed times, 5,000 and 15,000 years respectively, since the beginning of release. As shown in these figures, the concentration becomes closer to that from the solution without a limiting zone, as "*a*", the distance between the fracture and rock interface and an impermeable boundary, increases. When "*a*" is 5 meter (Fig. 2) or 10 meter (Fig. 3) then the two profiles are almost identical.

For smaller values of "*a*", the effect of a limiting capacity of matrix diffusion by the existence of an impermeable boundary becomes noticeable. These figures clearly show that if the real field data indicate that the so-called impermeable zone exists and its thickness is less than ten or five meters for non sorbing nuclides (in this case the retardation coefficient is one) then the current matrix diffusion models underestimate the overall safety.

Table 1 Input parameters for the numerical evaluations

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D_f	(m^2/yr)	1	v	(m/yr)	1.0
D_p	(m^2/yr)	0.01	$T_{1/2}$	(yr)	1.57×10^7
R_f	(-)	1	b	(m)	5.0×10^{-4}
R_p	(-)	1 (10, 10^2)	q	(-)	0.01
a	(m)	0.5,1.0,5.0,10.0	T	(yr)	10,000

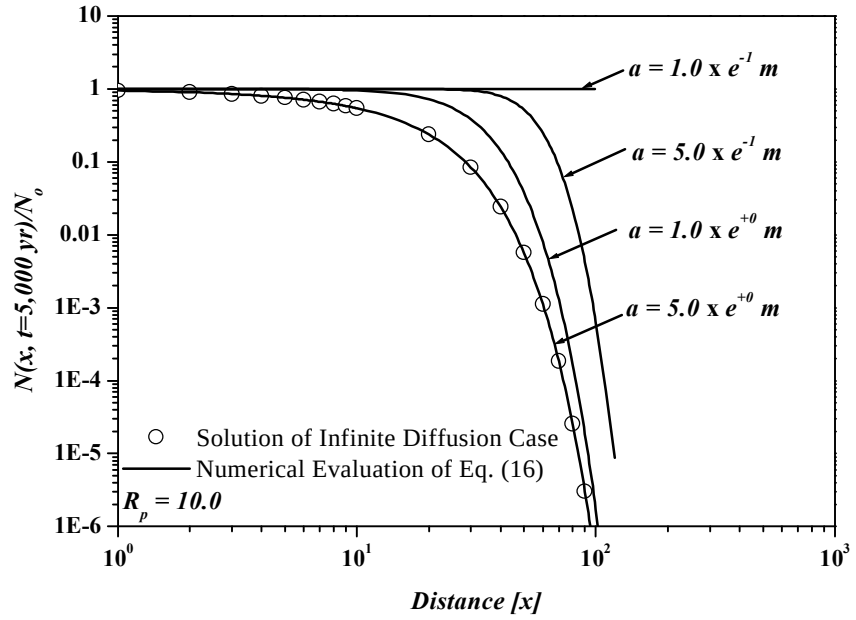

Figure 2 Normalized concentration with different thickness value, *a*, and the solution of infinite diffusion case [9], $R_p = 10.0$, $t = 5{,}000$ *yr*

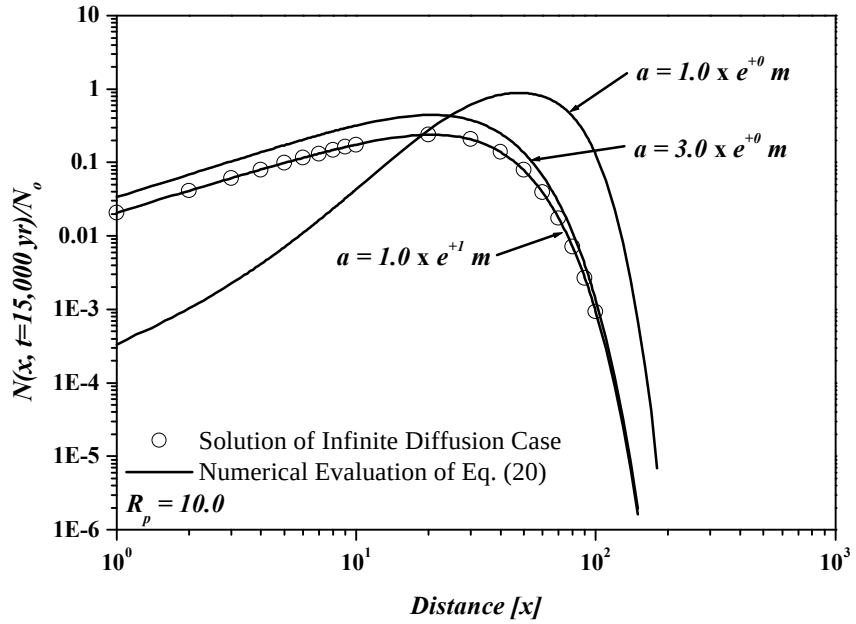

Figure 3 Normalized concentration with different thickness value, *a*, and the solution of infinite diffusion case [9], $R_p = 10.0$, $t = 15{,}000$ *yr*

5. Conclusions

Numerical inverse solutions are derived to estimate the concentrations of radionuclides in a fracture. The newly developed model was verified with the case of matrix diffusion for bigger "*a*" values. Results indicate that for smaller values of "*a*", the normalized concentrations deviate from those without the impermeable zone so that the current matrix diffusion models can not represent the real physical system accurately. Therefore, for conservative performance assessment, the newly developed model turns out to be more suitable to estimate concentrations in a fracture.

To fully assure the validity of this model, it is essential to find out the correct field data for this zone thickness and then check whether the existing rather simple solutions are good enough or the new, more complicated solutions in this paper are to be applied for safety assessment.

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