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A Numerical Model for Nuclide Migration in the Far-field of the Repository

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처분장 Far-field에서의 핵종이동 수치 모델

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Abstract

A numerical model for nuclide migration through fractured rock media has been developed. Nuclide transport with groundwater in rock fissures and the diffusion of nuclides into rock matrix are considered one-dimensionally. In the safety assessment of the repository for radioactive waste, this one-dimensional model by the finite-difference scheme, which enables us not only to use more realistic boundary conditions but also to model the nonhomogeneous rock medium as the multilayered media, can be used effectively with the analytical mode. The solution by the numerical model will be verified analytically, and then extended to the double-layered rock medium transport model.

요 약

중저준위 방사성 폐기물 처분 안전성 평가에 이용될 수 있는 유한 차분법에 의한 수치모델을 개발하였다. 이 모델은 처분장이 암반내에 위치한 경우에 대하여 처분장 주위 지하 암반매질에서의 핵종의 이동을 기술하는 것으로 암반내의 단일한 균열으로의 지하수에 의한 이동과 균열에 수직한 방향으로의 확산을 고려하여 일차원적으로 해석하였다. 수치모델은 해석해와 병행하여 처분장 안전성 평가에 있어서 유용하게 이용될 수 있는 것으로 보다 실제적인 경계조건을 사용할 수 있게 하고, 불균질한 암반매질에 대해 다중매질 모델링을 제공한다. 수치모델의 검증을 위하여 균열에서의 Sr-90 농도 Profile을 구하여 해석해와 비교하였고, 몇몇 경계 조건에 따른 영향을 비교하고 암반매질을 파라미터 값이 단계적으로 변하는 이중 매질에 대하여 확장하였다.

1. Introduction

It has been decided that low- and intermediate level radioactive wastes are to be disposed of in a repository constructed in the crystalline rock masses in Korea. This decision necessiates the prediction of behavior of the nuclides in rock media. Several well-known mathematical models for nuclide migration in the porous media at the far-field around the repository had been used in predicting the nuclide behavior before the 1980's. However, these models have some limitations related to the migration of nuclides through rock media. In crystalline rocks, such as granite and gneiss, it is known that nuclides are carried by groundwater, which flows mainly through the fissures. In most rock matrix, nuclides are considered to be transported by diffusion only.

Neretnieks[1] and *Grisak et al.*[2] proposed the coupled governing equations which describes the nuclide behavior both in the fissure and in the rock matrix where the single fissure splits off the porous rock matrix resulting in a "dual-porosity system". *Neretnieks*[1] analyusis of this model was based on a description of the conditions in a single fissure and on simple analytical solution for the case in which both dispersion and retardation in the fissure are neglected. *Grisak et al.*, obtained another solution by the finite-element method.

Recently, *Tang et al.*[3] developed transient as well as steady state analytical solutions with consideration of the dispersion in the fissure. since then this model has been extended by several authors. They have related specific microscopic features of the rock to several models.[4,5,6] All these models are based on an assumption that a part of the nuclides are diffused considerably into rock matrix orthogonally as transported through th fissure. This can enhance the retardation of the nuclides by some order of magnitude and contribute the radioactive decay.

In this study a numerical model for nuclide migration in the fissure and rock matrix is developed.

The numerical model can be applied variously to the estimation of radionuclide migration in the far-field when flux-type inlet and outlet boundary conditions must be considered. Flux-type boundary condition is a very necessary real situation in safety assessment of the repository.

In the analytical model it is very difficult to adopt various boundary conditions. Also, for the piecewise homogeneous multilayered rock media where some critical physicochemical parameter values are changed abruptly, the numerical model can be easily applied. In addition, using this model, the concentration profile of nuclides in the rock matrix as well as the fissure, can be obtained simultaneously. This cannot be done using the analytical model.

To this end, a model by a simple finite-difference scheme has been developed and verified by the analytical solution. This model is also extended to the double-layered medium transport model.

2. Transport Equation

A physical system is schematically represented in Fig. 1. It is assumed that if nuclides are released from the repository, then they are introduced directly to the inlet of the fissure. The permeability in rock matrix is so low that transport within it will be mainlky by molecular diffusion. Therefore transport along the fissure by advection and dispersion with groundwater is much faster than when nuclides are transported within the rock matrix. This fact provides the basis for taking the direction of mass flux in the rock matrix to be perpendicular to the fissure axis (z-axis). Therefore the two-dimensional system can be reduced to two orthogonal, coupled one-dimensional systems. As such simplified and amenable one-dimensional treatment for the fissure-rock matrix system becomes possible.

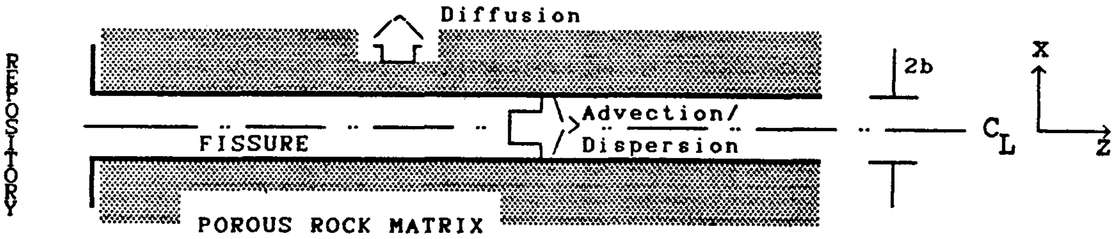

Fig. 1. Fissure-Rock Matrix System.

The processes to be considered here are advective transport, molecular diffusion and longitudinal mechanical dispersion in the fissure, diffusion from the fissure into rock matrix, adsorption onto the fissure surface and within the matrix, and radioactive decay.

The transport process in the system of Fig. 1 for the fissure-rock matrixk medium can be represented by two coupled differential equations.

For the fissure,

$$\frac{\partial C_f}{\partial t}=\frac{D_f}{R_f}\frac{\partial^2 C_f}{\partial Z^2}-\frac{V}{R_f}\frac{\partial C_f}{\partial Z}-\lambda C_f+\frac{\phi_a D_p}{bR_f}\frac{\partial C_p}{\partial X}\bigg|_{Z=b} \quad 0\leqq Z\leqq\infty \qquad (1)$$

where C_f is nuclide concentration in the fissure, 2b is fissure width, v is groundwater velocity, D_f is hydrodynamic dispersion coefficient expressed as $D_f=\alpha_L V+D^*$ where α_L is longitudinal dispersivity and D^* is molecular diffusion coefficient in water, λ is decay constant, and q is diffusive flux perpendicular to fissure axis. $R_f=1+K_a/b$ is the retardation coefficient on the fissure wall, K_a is the surface distribution coefficient, and ϕ_a is porosity of rock matrix.

For the rock matrix,

$$\frac{\partial C_p}{\partial t}=\frac{R_p}{D_p}\frac{\partial^2 C_p}{\partial X^2}-\lambda C_p \quad b\leqq X\leqq\infty \qquad (2)$$

where C_p is the nuclide concentration in the rock matrix, $R_p=1+\rho_b K_d/\phi_a$ retardation factor in the rock matrix, ρ_b is the bulk density of the rock, and K_d is the distribution coefficient. The boundary conditions for Eq. (1) are

$$C_f(Z,0)=0 \qquad (3a)$$
$$C_f(0,t)=C_0 e^{-\lambda t} \qquad (3b)$$
$$C_f(\infty,t)=0 \qquad (3c)$$

where C_o is te nuclide concentration in the repository, and Eq. (3*b*) represents constant leaching rate condition. The boundary condition for Eq. (2) are

$$C_p(X,Z,0)=0 \qquad (4a)$$
$$C_p(b,Z,t)=C_f(Z,t) \qquad (4b)$$
$$C_p(\infty,Z,t)=0 \qquad (4c)$$

Other boundary conditions are also considered for constant inlet flux boundary condition which corresponds to a constant flux vC_0 at $z=0$. For constant flux at the inlet of the fissure the flux-type Danckwerts' boundary condition can be represented as[7]

$$VC_0=\left(-D_f\frac{\partial C_f}{\partial Z}+VC_f\right)_{Z=0+} \qquad (5)$$

Also, for the outlet of the fissure,

$$\left.\frac{\partial C_f}{\partial z}\right|_{z=L} = 0 \tag{6}$$

where L is the length of the fissure.

The analytical solution to Eqs. (1) and (2) for the concentration of the fissure can be easily obtained as Eq. (7)[8].

$$C_f(Z,t) = \frac{2C_0 \exp\{vz/(2D_f)\}}{\pi^{1/2}} \exp(-\lambda t) \int_l^\infty \exp\left[-\xi^2 - \frac{v^2 z^2}{16\xi^2 D_f^2}\right] d\xi$$
$$\times \operatorname{erfc}\left\{\frac{Z^2 \phi_d D_p (D_p R_p)^{1/2}}{8\xi^2 D_f b} \left(t \frac{Z R_f^2}{4\xi^2 D}\right)^{-1/2}\right\} d\xi$$

where $l = Z/2 \cdot (R_f/D_f t)^{1/2}$

3. Numerical Solution

The region of interest is represented by the grid points as shown in Fig. 2. Both of the fissure and the rock matrix will be considered one-dimensionally.

Once C_{f_i} is obtained, then the new $C_{p_{i,l}}$ can also be obtained in the direction perpendicular to the fissure one-dimensionally with the new inlet boundary condition for rock matrix, which is given as the C_{f_i}. So, C_{f_i} can be obtained again at that time level by iterating scheme.

Eq. (1) is replaced by the finite-difference equation for $1 \leqslant i \leqslant N\text{-}1$ by the central difference scheme.

$$(\alpha-\beta)\vartheta\, C_{f_{i+1}}^{\nu+1} + [2(1+\beta\vartheta) + \gamma\vartheta]C_{f_i}^{\nu+1} - (\alpha+\beta)\vartheta\, C_{f_{i-1}}^{\nu+1}$$
$$= -(\alpha-\beta)(1-\vartheta)\, C_{f_{i+1}}^{\nu} + [2\{1-(1-\vartheta)\beta\} - \gamma(1-\vartheta)]\, C_{f_i}^{\nu}$$
$$+ (\alpha+\beta)(1-\vartheta)\, C_{f_{i-1}}^{\nu} + \gamma\vartheta\, C_{p_{i,1}}^{\nu+1} + \gamma(1-\vartheta)\, C_{p_{i,1}}^{\nu} \tag{8}$$

where $\alpha = \Delta t \cdot V/(R_f \cdot \Delta Z)$, $\beta = 2\Delta t \cdot D_f/\{R_f(\Delta Z^2)\}$, $\gamma = \phi_d \cdot D_p \cdot \Delta t\,(bR_f \cdot \Delta X)$, ν refers to previous time level, whereas $\nu+1$ refers to the next time level and ϑ is the weight parameter, which is 1/2 for the

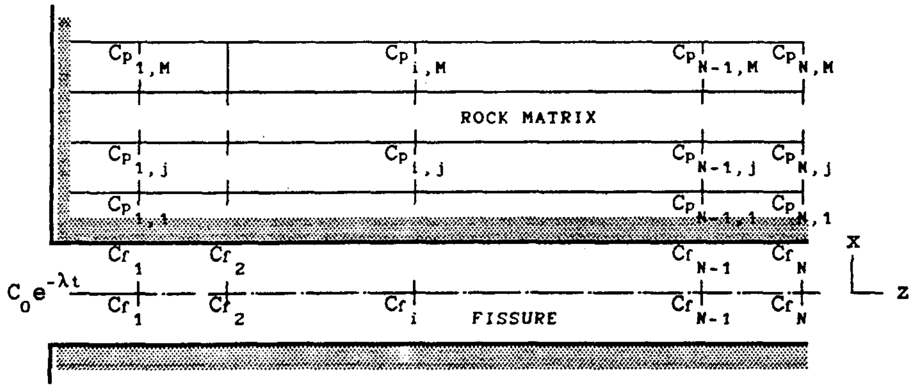

Fig. 2. Schematic modeled region showing finite-difference discretization.

Since Eqs. (1) and (2) are coupled with each other, numerical solutions are obtained by marching forward in time, solving the simultaneous set of difference equations at each time level. This is done in an iterative manner. First, the concentration in the fissure, C_{fi} is found for one-dimensional grids, i, with the assumtion that the concentration in the rock matrix is $C_{pi,l}$.

Crank-Nicolson scheme. In case of the flux inlet boundary condition, for grid $i=0$, from Eq. (8),

$$-2\beta\vartheta\, C_{f_1}^{\nu+1} + [\xi + (\alpha+\beta)\vartheta\varphi]\, C_{f_0}^{\nu+1}$$
$$= [2\beta(1-\vartheta)\varphi]\, C_{f_1}^{\nu} + [\zeta - (\alpha+\beta)(1-\vartheta)\psi]C_{f_0}^{\nu} + (\alpha+\beta)\varphi\, C_0$$
$$+ \gamma\vartheta\, C_{p_{0,1}}^{\nu+1} + (1-\vartheta)\, C_{p_{0,1}}^{\nu} \tag{9}$$

where $\xi=2(1+\beta\vartheta)+\gamma\vartheta$, $\zeta=2[1-\beta(1-\vartheta)]-\gamma(1-\vartheta)$, and $\varphi=2\cdot\Delta z\cdot v/D_f$.

For the Danckwerts' type outlet boundary condition, for grid $i=N$,

$$[2(1+\beta\vartheta)+\gamma\vartheta]C_{f_N}^{\nu+1}-2\beta\vartheta\, C_{f_{N-1}}^{\nu+1}$$
$$=2\beta(1-\vartheta)\,C_{f_{N-1}}^{\nu}+[2\{1-(1-\vartheta)\beta\}-\gamma(1-\vartheta)]\,C_{f_N}^{\nu}+\gamma\vartheta\,C_{p_{N,1}}^{\nu+1}+\gamma(1-\vartheta)\,C_{p_{N,1}}^{\nu} \quad (10)$$

For the rock matrix, finite-difference equation can be obtained similarly.

4. Verification fo Numerical Solution

For the numerical model, the Crank-Nicolson, the centered-space, finite-difference scheme is used. It is well known that the advective-dispersion equation has many problems of oscillatory solution and numerical dispersion. The problems are particularly severe when advection dominates over dispersion. The numerical oscillation of solution tends to be more severe as the front of the concentration profile becomes sharper. Also, numerical dispersion stems primarily from first-order finite-difference approximation to the first-order time and space derivatives in Eq. (1). Consequently, at least second-order approximations must be considered. Numerical dispersion leads to smearing of the concentration front. Selecting an appropriate time and space grid step size can minimize the oscillatory solution and compensate for the artificial numerical dispersion coefficient for the physical dispersion coefficient. In this study. however, in the case of the centered, Crank-Nicolson scheme no numerical dispersion is appeared. The numerical model must be compared to analytical solution to establish its accuracy. To this end a test calculation of concentration profile for the nuclide, Sr^{90} is done. Table 1 gives some input parameter values, which are adopted from a variety of literature.[9,10,11]

Table 1. Input parameter values.

Parameter	Value
Nuclide	Strontium-90
half-life	29 [year]
K_d	1.7 [cm³/g]
K_a	7.0×10^{-3} [m]
Half-width of the fissure, b	0.0011 [m]
Porosity, ε_p	0.005
Tortuosity factor, τ	0.1
Molecular diffusivity, D^*	0.05 [m²/year]
Rock bulk density, ρ_b	2.62 [g/cm³]
Dispersion length, α_L	0.1 [m]
Groundwater velocity, v	10 [m/year]
Length of fissure	1 [m]

Fig. 3 shows the comparison of two concentration profiles of Sr^{90} calculated by the analytical and the numerical models, respectively. Agreement is good. For sensitivity of parameters, the behaviors of the concentration profiles of Sr^{90} are represented in Figs. 4a and 4b. They agree well with each other when parameters such as half-width of the fissure, groundwater velocity, diffusion coefficient in the rock matrix, K_d, and K_a are increased by a factor of two for the base case calculation with parameter values represented in Table 1.

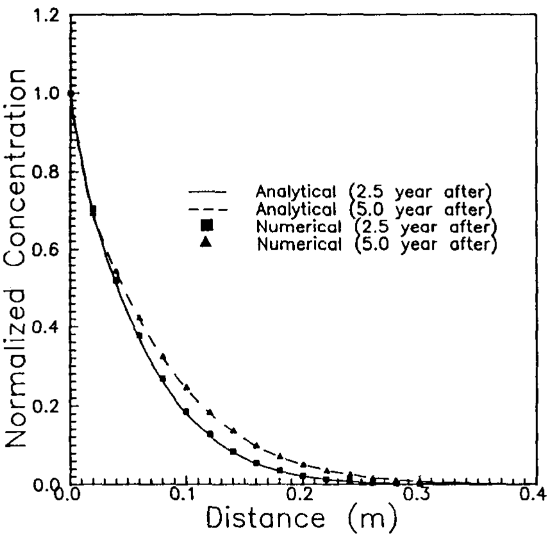

Fig. 3. Comparison of numerical solutions with analytical solutions for Sr^{90} after 2.5 and 5.0 years.

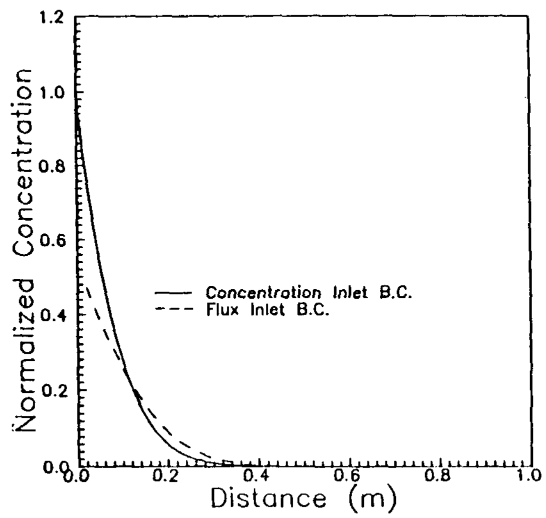

Fig. 5. Effect of inlet boundary condition on the concentration profile in the fissure.

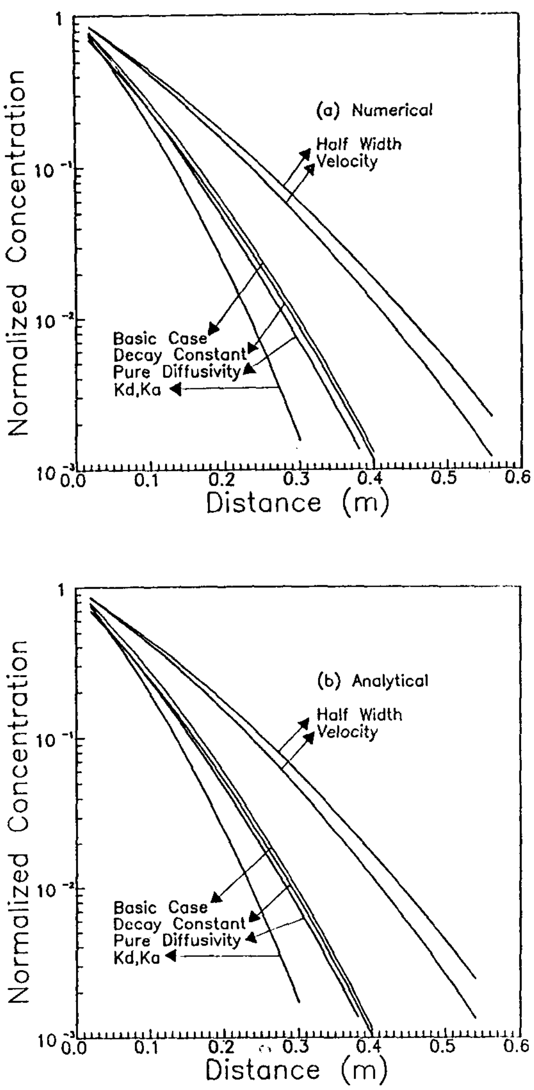

Fig. 4. Effect of some parameter values on the concentration profile in the fissure: (*a*) Numerical solution and (*b*) Analytical solution.

5. Application of the Numerical Model

According to INTRACOIN study,[12] an international cooperation project for comparing models for transport of nuclides in geologic media, one of the causes of deviating results of the numerical calculation for various model is the use of different boundary condition even for the same models. Using appropriate boundary conditions is very important in view of the relationship of the far-field model with other near-field models and in light of the quantitative effect of the boundary condition. However, the analytical solution is available only for the limited boundary condition types.

To show the impact of the boundary condition on the concentration profiles in the fissure, a profle with constant concentration boundary condition is compared in Fig. 5 with the profile assuming constant flux boundary condition. As is seen, the concentration obtained with constant concentration boundary condition is much different from that with constant flux boundary condition.

Moreno et al.[13] derived the analytical solution for constant inlet flux boundary condition. It is known that the concentration profile in the rock matrixk is strongly influenced by the inlet boundary condition type. Flux-type boundary conditions at the inlet can be used when the flux of nuclide calculated from the source-term model and/or when the near-field diffusion model is used as the input for far-field model and such boundary conditions suitable in real situation in the safety assessment of the repository.

To predict the quantity of the nuclide diffused into the rock matrix, two models are considered separately and compared with each other. The first model assumes that the nuclide is transported through the fissure without any diffusion into the rock matrix, whereas the other implies that the nuclide is transported through both the fissure and the rock matrix.

Concentration profiles produced by the numerical solution and the analytical solution,[14] respectively are compared with the first model and agreement is good.

Fig. 6 shows a schematic three-dimensional concentration profile in the fissure and the rock matrix as a example.

Another concentration profile in the fissure for various molecular diffusion coefficients is represented in Fig. 7, from which we know that if D_p increases, then the diffusion into the rock matrixk becomes more dominant.

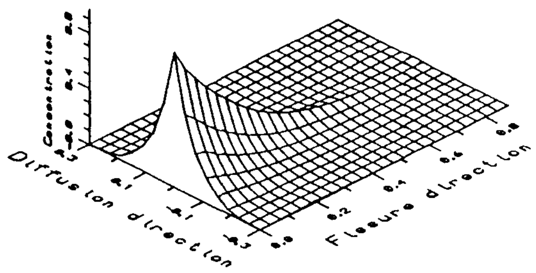

Fig. 6. Schematic three-dimensional concentration profile.

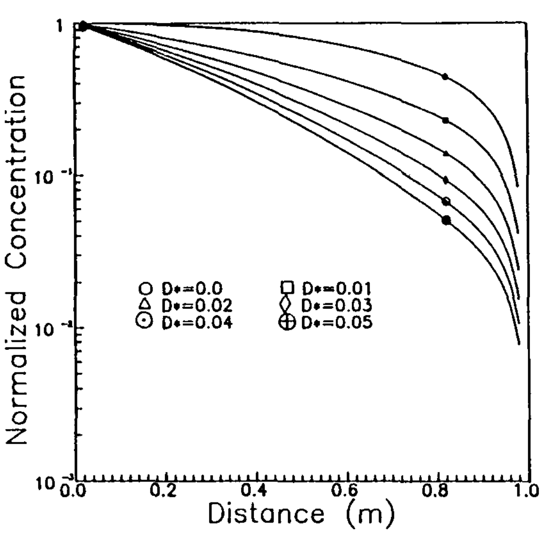

Fig. 7. Effect of diffusion coefficient in the rock matrix on the concentration profile in the fissure.

6. Double-Layered Media Model

For the different layers which are assumed to be characterized by piecewise constant parameters, a numerical model is also developed.

In Fig. 2, another medium prolonged beside $i=N$, which has grid point from $i=N+1$ to $i=L$, can be considered. With additional boundary conditions at the interlayer according to the flux and concentration continuity conditions are specified as[12,15]

$$b_1\left[D_f^1\frac{\partial C_f^1}{\partial z}-v_1C_f^1\right]_{z=l_1-0}=b_2\left[D_f^2\frac{\partial C_f^2}{\partial z}-v_2C_f^2\right]_{z=l_1+0} \tag{11}$$

$$C_{f_N}^{1,\langle\nu\rangle}=C_{f_N}^{2,\langle\nu\rangle}=C_{f_N}^{*,\langle\nu\rangle} \tag{12}$$

where l_l is the length of the layer 1, and sub- and superscript, 1 and 2 represent the layer 1 and the layer 2, respectively.

With Eqns. (11) and (12), applying the central difference scheme, the difference equation at the interlayer, $i=N$ is obtained as

$$\begin{aligned}&\tau\vartheta\, C_{f_{N+1}}^{2,\nu+1}+\{(b_2v_2-b_1v_1-\sigma-\tau)\vartheta\}\, C_{f_N}^{*,\nu+1}+\sigma\vartheta\, C_{f_{N-1}}^{1,\nu}\\&=\{-\tau(1-\vartheta)\}\, C_{f_{N+1}}^{2,\nu}+\{(\sigma+\tau+b_1v-b_2v_2)(1-\vartheta)\}\, C_{f_N}^{*,\nu}\\&+\{-\sigma(1-\vartheta)\}\, C_{f_{N-1}}^{1,\nu}\end{aligned} \tag{13}$$

where $\sigma=b_1D^1/\Delta z$, $\tau=b_2D^2/\Delta z$.

Finite-difference equations for double-layered media can then be represented as in the following matrix form.

$$\begin{pmatrix}\{2(1+\beta\vartheta)+\gamma\vartheta\} & \vartheta(\alpha-\beta) & & & & & \\ -(\alpha+\beta)\vartheta & \{2(1+\beta\vartheta)+\gamma\vartheta\} & \vartheta(\alpha-\beta) & & \emptyset & & \\ \ddots & \ddots & \ddots & & & & \\ & -(\alpha+\beta)\vartheta & \{2(1+\beta\vartheta)+\gamma\vartheta\} & \vartheta(\alpha-\beta) & & & \\ & & (\sigma\vartheta) & (b_2v_2-b_1v_1-\sigma-\tau) & (\tau\vartheta) & & \\ & & & -(\alpha'+\beta')\vartheta & \{2(1+\beta'\vartheta)+\gamma'\vartheta\} & \vartheta(\alpha'-\beta') & \\ & & & \ddots & \ddots & \ddots & \\ \emptyset & & & & -(\alpha'+\beta')\vartheta & 2(1+\beta'\vartheta)+\gamma'\vartheta & \vartheta(\alpha'-\beta') \\ & & & & & -(\alpha'+\beta')\vartheta & 2(1+\beta'\vartheta)+\gamma'\vartheta\end{pmatrix}\begin{pmatrix}C_f^1\\C_f^2\\\vdots\\C_f\\C_f^*\\C_f\\\vdots\\C_f\\C_f\end{pmatrix}^{\nu+1}=\begin{pmatrix}R\end{pmatrix}^{\nu,1} \tag{14}$$

where $\alpha=\Delta tv_1/(R_f^1\cdot\Delta z_1)$, $\beta=2D_f^1\Delta t/\{R_f^1(\Delta z_1)^2\}$, $\gamma=\phi_d^1D_p^1\Delta t/b_1R_f^1\Delta x$, $\alpha'=\Delta tv_2/(R_f\cdot\Delta z_2)$, $\beta'=2D_f^2\Delta t/\{R_f^2(\Delta z_2)^2\}$, $\gamma'=\phi_d^2D_f^2\Delta t/b_2R_f^2\Delta x$, and R is the constant vector term.

According to *Choi et al.*[16] fissure width is very sensitive for the behavior of the concentration profile in the fissure.

Fig. 8 shows the behavior of the Sr^{90} concentration profile for various couples of half-width of the Fissure. The behavior of the concentration profile is shown in Fig. 9 when several parameters such as K_d, K_a, porosity in the rock matrix, and diffusivity are varied by a factor of two.

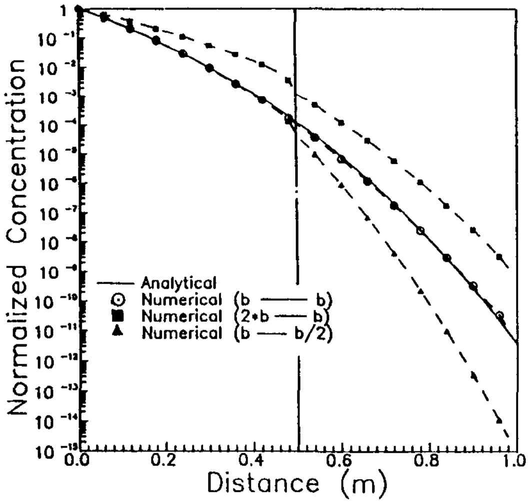

Fig. 8. Effect of half-width of the fissure on the concentration profile in the fissure.

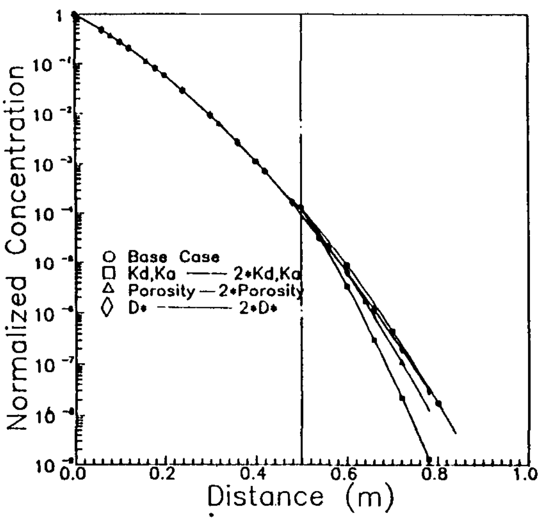

Fig. 9. Effect of some parameters in the fissure on the concentration profile.

7. Conclusion

In the present study a numerical model describing the nuclide migration through fractured rock media has been developed and verified. Because analytical solution has many drawbacks in safety assessment, the numerical model may be applicable where the analytical model is limited due to the boundary condition and the connection with other models. In addition, the numerical model is applicable to the multilayered media which has the piecewise constant parameter values. These numerical models agree well with the analytical models and behave well with the variation of parameter values.

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