Analytical Solutions for a Three-Member Decay Chain of Radionuclides Transport in a Single Fractured Porous Rock

Young Woo Yu and Chang Hyun Chung

Seoul National University

Chang Lak Kim

Korea Atomic Energy Reaserch Institute

(Received October 22, 1992)

단일균열 다공성암반에서 방사성핵종의 수송에 대한 3단계 붕괴사슬의 해석해

유영우 · 정창현

서울대학교

김창락

한국원자력연구소

(1992. 10. 22 접수)

Abstract

The migration equation is modified for a three-member decay chain in the fracture and porous matrix. Analytical solutions are obtained by utilizing Laplace transform the initial conditions of Delta function and Bateman equation.

The concentrations for each nuclide of Np^{237}-U^{233}-Th^{229} and U^{234}-Th^{230}-Ra^{226} chains selected from the $4n+1$ and $4n+2$ chains are plotted by utilizing analytical solutions in the fracture. Retardation coefficients of the nuclides are obtained using those of the granite.

The results indicate that the daughter nuclides such as U^{233}, Th^{229}, Th^{230} and Ra^{226} become important at the far field from the repository though there is very small initial inventory in the waste solid or spent fuel, for they are produced by the mother nuclides decayed in the fracture and porous matrix.

요 약

암반(Porous Rock Matrix)과 균열(fracture)에서 일차원의 이동 방정식(Migration Equation)을 3-Member Decay Chain까지 확장하고, Laplace Transform을 이용하여 초기조건이 Delta Function 과 Bateman Equation인 각각에 대해 해석해를 구한다. 그 해를 이용하여 Actinide Chain 중 4n+1과 4n+2 Chain에서 선택된 Np^{237}-U^{233}-Th^{229}와 U^{234}-^{230}-Ra^{226} chain의 각 핵종들의 균열에서의 농도를 상대농도로 나타낸다. 이핵종들의 지연계수(Retardation Coefficient)는 화강암에 대한 것을 사

용하여 균열에서의 농도 변화를 볼 수 있다.

본 연구에 의한 결과로는 U^{233}, Th^{229}, Th^{230}, Ra^{226} 같은 핵종들은 비록 초기 inventory에는 작은 양일지라도 균열과 암반에서 모핵종의 붕괴(decay)에 의해 생기므로써 처분장으로부터 먼 거리에서는 중요한 핵종이 된다는 것을 알 수 있다.

1. Introduction

Most of radioactive wastes generated from nuclear power plants will remain radioactive for a very long time. The dense geological formations of radioactive waste repository are especially important for confining relatively long half-life nuclides. As radionuclides dissolve in groundwater among the dense geological formation effects, it is essential in predicting the safety of the geological disposal system to understand how radionuclides are transported into and through geological layser.[1]

The prime requirements for rock matrix of the waste repository are low permeability and porosity. In fractured porous media, all of the groundwater flow occur within the fractures because fractures have permeabilities of several orders of magnitude larger than those of the rock matrix, if the geological layers are fully saturated with water. So radionuclides dissolved in groundwater will be transported along a fracture with molecular diffusion from the fracture to the rock matrix.[2]

Molecular diffusion from the fractures into the porous matrix constitutes an attenuation mechanism that can be highly effective in removing contaminant mass from the primary flow channels and thus in retarding the advance of contaminant in the system. In the case of a radioactive contaminant having a constant source strength, the advance will eventually cease if the flow is sufficiently long. The distribution of the contaminant in the system will be stable if the effect of the daugthers generated by the parent species is neglectd due to the loss by decay of mass stored in both the fractures. And the porous matrix will balance the mass input at the source and that due to decay of parent species.

In practice radioactive wastes have multi-decay chains, the daughter nuclides produced by the mother nuclide will be increased along the fracture though there is very small initial inventory in the waste solid or spent fuel. Neglecting radioactive-decay precursors affects in two ways. First, total amount of a daughter radionuclide in the source and in the medium would be underestimated which results in underestimation of maximum concentration. Second, if the mother nuclide is transported faster, the daughter nuclides would exist in a extended region than expected because the daughter is generated by the mother nuclide decayed in the fracture and in the porous matrix.

Analytical solutions for the transport of a two-member decay chain have been developed by Sudicky and Frind[2] for one-dimensional transport along a single at a porous rock matrix. Joonhong Ahn[1] obtained analytical solutions for a two member decay chain as Green's functions for the boundary conditions prescribed by an arbitrary function of time. Actinides chains in high level wastes practically have more than two members and cannot be treated by the method above. Since some elements have very short half lives while some others long half-lives, these chains can be broken into shorter chains.[3]

U^{234} from the 4n+2 chain in spent fuel has decayed significantly, the contributions of Th^{230} and Ra^{226} comes from Am^{242}, and all remaining nuclides of the cases treated in this work are in secular equilibrium with Ra^{226}. On the other hand, there is a large amount of Pu^{241}. For practical purpose, the quantity of nuclides chain which is beginned with decay of Pu^{241} to form Am^{241} and Np^{237} is not a little. If waste dissolution begins hundreds of years or more after emplacement, the 4n+1 chain can be reduced

to the following chain: Np^{237}-U^{233}-Th^{229} where the initial amount of Np^{237} should include the decay of Pu^{241} and Am^{241}, because the half lives of Pu^{241} and Am^{241} are shorter. Therefore, Np^{237}-U^{233}-Th^{229} and U^{234}-Th^{230}-Ra^{226} chains are selected among the same sequence, the four distinct complete actinide chains present in high level wastes can be broken into the three-member decay chains.

Matrix diffusion and chemical sorption of multi-decay chains will be predicted indirectly through the analytical solutions for a three-member decay chain. This paper solves migration equations of a three-member decay chain for one-dimensional transport through a single fracture. The analytical solutions are obtained by analytical inversion of Laplace transform, the numerical calculation of the analytical solutions is utilized by the trapezoid method because the analytical solution forms are expressed in the integral forms.

2. Theory

2.1. Assumptions

In order to permit one dimensional analysis of transport along the fracture for one dimensional molecular diffusion of radionuclides from the fracture to the porous matrix, we will make several assumptions pertaining to the geometric and hydraulic properties of the system at hand; These assumptions are; the width of the fracture is much smaller than its length; transport in the porous rock matrix is controlled by molecular diffusion because the intack rock has low hydraulic conductivity; transport along the fracture by advection is much more rapid than transport in the rock matrix; longitudinal dispersion in a fracture is neglected; a groundwater velocity along the fracture is constant[2]

2.2. The Physical Process

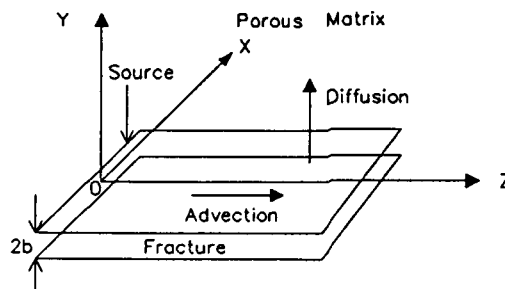Consider a porous matrix containing one

Fig. 1. Geometry and Physical Processes Considered in the Model With a one Dimensional Single Fracture.

dimensional single fracture of width 2b, situated in a water-saturated porous rock of porosity ε.(See Figure 1)

The transport of radioactive contaminants in the fracture and porous rock matrix will be formulated with consideration given to the following process:a) Advective transport at a constant groundwater velocity along the fracture. b) Molecular diffusion from the fracture to the porous matrix. c) Sorption onto the fracture surfaces. d) Sorption within the porous rock matrix. e) Radioactive decay with 3 member precursors in the fracture and porous rock matrix with decay constant λ.[2]

2.3. Governing equations

The transport of each member of 3-member decay chain can be described by three coupled one dimensional equations on the basis of the above assumptions; one set for the fracture and one set for the porous rock matrix.

The coupling here is provided by the continuty of concentration along the fracture-matrix interface. Longitudinal dispersion in a fracture is neglected.

1) Governing equatons for one dimensional transport at a fracture:

$$R_{f1}\frac{\partial N_1}{\partial t} + V\frac{\partial N_1}{\partial z} + \lambda_1 R_{f1}N_1 + \frac{q_1}{b} = 0$$

$$(z > 0, \quad t > 0) \quad (2.1)$$

$$R_{f2} \frac{\partial N_2}{\partial t} + V \frac{\partial N_2}{\partial z} + \lambda_2 R_{f2} N_2$$

$$- \lambda_1 R_{f1} N_1 + \frac{q_2}{b} = 0$$

$$(z > 0, \quad t > 0) \quad (2.2)$$

$$R_{f3} \frac{\partial N_3}{\partial t} + V \frac{\partial N_3}{\partial z} + \lambda_3 R_{f3} N_3$$

$$- \lambda_2 R_{f2} N_2 + \frac{q_3}{b} = 0$$

$$(z > 0, \quad t > 0)(2.3)$$

N_i: Concentration in a fracture

2) Governing equatons for one dimensional transport at a porous matrix:

$$R_{p1} \frac{\partial M_1}{\partial t} - D_{p1} \frac{\partial^2 M_1}{\partial y^2} + \lambda_1 R_{p1} M_1 = 0$$

$$(y > b, \quad z > 0, \quad t > 0)(2.4)$$

$$R_{p2} \frac{\partial M_2}{\partial t} - D_{p2} \frac{\partial^2 M_2}{\partial y^2} + \lambda_2 R_{p2} M_2 - \lambda_1 R_{p1} M_1 = 0$$

$$(y > b, \quad z > 0, \quad t > 0) \quad (2.5)$$

$$R_{p3} \frac{\partial M_3}{\partial t} - D_{p3} \frac{\partial^2 M_3}{\partial y^2} + \lambda_3 R_{p3} M_3 - \lambda_2 R_{p2} M_2 = 0$$

$$(y > b, \quad z > 0, \quad t > 0)(2.6)$$

M_i: Concentration in a porous matrix
Subscripts 1, 2, 3 in the above equations stand for mother and daughter muclides, respectively.

3) $q_i(z, t)$ are defined as:

$$q_i(z,t) = - \varepsilon D_{pi} \frac{\partial M_i}{\partial y} \Big|_{y=b}$$

$$(z > 0, \quad t > 0) \quad i=1,2,3 \quad (2.7)$$

The loss term q_i in the fracture equations represents the diffusive flux crossing the fracture-matrix boundary.
where

ε: porosity

$D_{pi} = \tau D_i$ (τ: tortuosity of the rock matrix

D_i: free solution molecular diffusion coefficient)

D_{pi}: molecular diffussion coefficient in the porous rock matrix

$R_{fi} = 1 + \frac{K_{fi}}{b}$: retardation coefficient in the fracture

$R_{pi} = 1 + \frac{\rho_b}{\varepsilon} (K_p)_i$: retardation coefficient in the porous matrix

K_{fi}: sorption distribution coefficient in the fracture (L)

K_{pi}: sorption distribution coefficient in the porous rock matrix (L^3/M)

ρ_b: bulk density of the porous matrix (L^3/M)

$\lambda_i = \frac{\ln 2}{(t_{1/2})_i}$: decay constant

4) Side conditions

N_i of the following conditions are imposed on the system of equations describing transport in the fracture. M_i of the following conditions are imposed on the system of equations describing transport in the porous rock matrix. $N_i(0, t)$ condition is expressed in the form of Delta function and Bateman equation. $N_i (z, t) = M_i(b, z, t)$ condition is interface condition of surface of fracture-porous rock matrix.

$$N_i(z,0) = 0 \qquad\qquad (z > 0) \qquad\qquad (2.8)$$

$$M_i(y,z,0) = 0 \qquad (y > b, \ z > 0)(2.9)$$

$$N_i(0,t) = \Psi_i(t) \qquad (t > 0) \qquad (2.10)$$

$$N_i(\infty,t) = 0 \qquad\qquad (t > 0) \qquad\qquad (2.11)$$

$$N_i(z, t) = M_i(b, z, t) \quad (z > 0, \ t > 0) \quad (2.12)$$

$$M_i(\infty, z, t) = 0 \quad (z > 0, \ t > 0) \quad (2.13)$$

$$i = 1, 2, 3$$

3. Solutions and Results

3.1. Solutions

The solutions are obtained from two initial conditions of Delta function and Bateman equation. The case 1 represents solutions from the initial condition of Delta function. The case 2 represents solutions from the initial condition of Bateman equation with band theory. Physical meaning of the case 1 is the concentration of the radionuclide that is deposited instantaneously at the coordinate origin of the fracture.[2] Physical meaning of the case 2 is the concentration of the radionuclide that is deposited continuously with radionuclides decayed from the source during the leach time.[4]

The Laplace transform method is utilized to solve equations for the fracture and the porous rock matrix with their respective boundary and initial conditions. The paper omits to process in detail the solving procedure and represents only solution parameter. Solving procedure is given as follows: 1) Laplace transform the equation which describes diffusive transport of the first member of the decay chain in the porous matrix. 2) Solve diffusive transport of the first member of the decay in porous matrix with Laplace transformed boundary conditions. 3) The transformed concentration gradient at the interface y=b is easily obtained by differentiation of the first member transformed solution of the decay chain in the porous matrix. 4) Laplace transform the equation which describes the transport of the first member of the decay chain in the fracture. 5) Solve transport of the first member of thedecay chain in the fracture with the transformed interface gradient and boundary

conditions in the fracture.[5] 6) Repeat the procedure from 1) to 5) with two and three member decay chain. 7) Inverse Laplase transformed solutions.[6]

Only here, the solution for the case 2 is represented. As for the case 1, refer to the original work.[7]

The $\Psi_i(t)$ are obtained form the Bateman equation with the band release fora 3-member decay chain as follow:

$$\Psi_1(t) = N_1{}^0 \exp(-\lambda_1 t) \ \{h(t)-h(t-T)\},$$

$$\Psi_2(t) = \left[N_2{}^0 \exp(-\lambda_2 t) \right.$$

$$\left. + \frac{N_1{}^0 \lambda_1}{(\lambda_2-\lambda_1)} [\exp(-\lambda_1 t)-\exp(-\lambda_2 t)] \right]$$

$$\{h(t)-h(t-T)\},$$

$$\Psi_3(t) = \left[N_3{}^0 \exp(-\lambda_3 t) + \frac{N_2{}^0 \lambda_2}{(\lambda_3-\lambda_2)} [\exp(\right.$$

$$-\lambda_2 t)-\exp(-\lambda_3 t)]+N_1{}^0 \lambda_1 \lambda_2 \left[\right.$$

$$\left[\frac{\exp(-\lambda_1 t)}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)} + \frac{\exp(-\lambda_2 t)}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)} \right.$$

$$\left. \left. + \frac{\exp(-\lambda_3 t)}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)} \right] \right] \ \{h(t)-h(t-T)\},$$

where $N_i{}^0$ is the concentration of the ith radionuclide that is deposited instananeously at the coordinate origin of the fracture.

Analytical solution from the initial condition of Bateman equation can be expressed by applying the convolution theorem with respect to time as follow:

$$N_i(z, t) = \int_0^t \Psi_i(t-\tau) \ \Psi_i(b, z, \tau) d\tau$$

$$(z \geq 0, \ t \geq 0) \quad (3.1)$$

$$M_i(y, z, t) = \int_0^t \Psi_i(t-\tau) \Psi_i(y, z, \tau) d\tau$$

$$(y \geq b, \quad z \geq 0, \quad t \geq 0) \quad (3.2)$$

3.2. Results

$$N_2(y, z, t) = \int_0^t \Psi_1(t-\tau) \, U_{12}(b, z, \tau) d\tau$$

$$+ \int_0^t \Psi_2(t-\tau) W_2(b, z, \tau) d\tau$$

$$(z \geq 0, \quad t \geq 0) \, (3.3)$$

$$M_2(y, z, t) = \int_0^t \Psi_1(t-\tau)\{U_{12}(b, z, \tau)$$

$$+ X_1(y, z, \tau)\} d\tau$$

$$+ \int_0^t \Psi_1(t-\tau) W_2(y, z, \tau) d\tau$$

$$(y \geq b, \quad z \geq 0, \quad t \geq 0) \, (3.4)$$

$$N_3(z, t) = \int_0^t \Psi_1(t-\tau)\{K_1(b, z, \tau)$$

$$+ K_2(b, z, \tau)\} d\tau$$

$$+ \int_0^t \Psi_2(t-\tau) U_{23}(b, z, \tau) d\tau$$

$$+ \int_0^t \Psi_3(t-\tau) W_3(b, z, \tau) d\tau$$

$$(z \geq 0, \quad t \geq 0) \quad (3.5)$$

$$M_3(y, z, t) = \int_0^t \Psi_1(t-\tau)\{K_1(y, z, \tau)$$

$$+ K_2(y, z, \tau) + X_2(y, z, \tau)$$

$$+ X_3(y, z, \tau)\} d\tau + \int_0^t \Psi_2(t-\tau) U_{23}(y, z, \tau) d\tau$$

$$\int_0^t \Psi_3(t-\tau) W_3(y, z, \tau) d\tau$$

$$(y \geq b, \quad z \geq 0, \quad t \geq 0) \quad (3.6)$$

The numerical calculation of the analytical solutions is performed through the trapezoid method.[7] The integral ranges with the given time are subdivided into about 2,000,000 intervals near field from the source, for the values of the given solutions change very sensitively at near field from the source. And the integral ranges are subdivided into about 100,000 far field from the source. The leach time is 30,000 yr. The retardation coefficients of each nuclides selected are taken from those of the granite. The results of numerical calculation are only represented forth case 2 in the fracture.

In Figure 2, profiles of concentration in the fracture for a chain, Np^{237}-U^{233}-Th^{229}, at 10,000 yr and 50,000 yr with $R_{p237} = 100$, $R_{p233} = 50$ and $R_{p229} = 5$,000 are plotted, where the leach time 30,000 yr in the fracture is considered in the latter calculation. The concentrations for U^{233} over 2,000m at 10,000 yr and over 6,000m at 50,000 yr are greater than those of the mother nuclide, for retardation coefficient of the daugther is less than that of the mother. The concentrations for Th^{229} over 200m at 10,000 yr and over 400m at 50,000 yr are greater decayed ones in the fracture than in the repository.

Also, profiles of concentration in the fracture for a chain, U^{234}-Th^{230}-Ra^{226}, at 10,000 yr and 50,000 yr (leach time 30,000 yr) with $R_{p234} = 50$, $R_{p226} = 500$ are plotted in the figure 3.

4. Discussions and Conclusions

The solutions for a 2-member decay chain contained several incorrect expressions. That method was able to express correct analytical solutions for a 2-member decay chain. The solution forms were not expressed as an Error function because of convenience for integral numerical calculation. Although the analytical solution was currently restricted to a 3-member decay chain, it could predict indirectly for
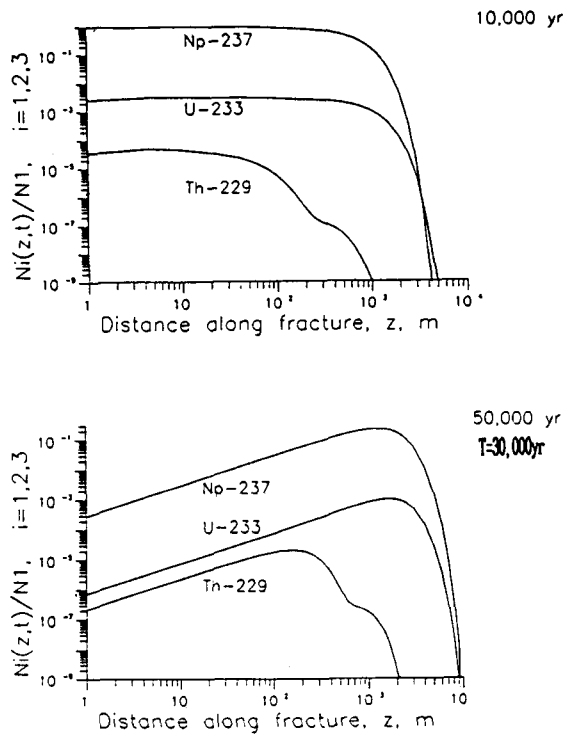
Fig. 2. Concentration Profiles of a Np^{237}-U^{233}-Th^{229} Chain From the Initial Condition of Bateman Equation in the Fracture.
($2b$=0.01m, v=10m/yr, ε=0.01, N^0_{237}=1, N^0_{233}=N^0_{229}=0, $R_{\varrho 237}$=$R_{\varrho 233}$=$R_{\varrho 229}$=1, R_{p237}=100, R_{p233}=50, R_{p229}=5,000 D_{p233}=D_{p229}=0.01m²/yr)
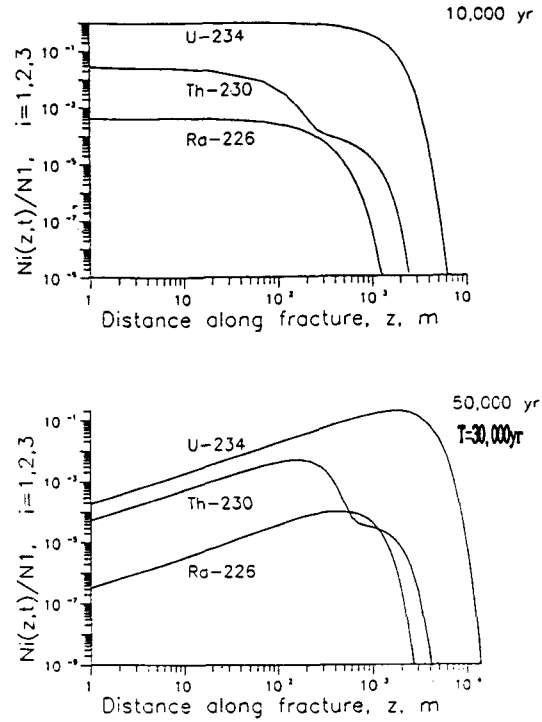

Fig. 3. Concentration Profiles of a U^{234}-Th^{230}-Ra^{226} Chain From the Initial Condition of Bateman Equation in the Fracture.
($2b$=0.01m, v=10m/yr, ε=0.01, N^0_{234}=1, N^0_{230}=N^0_{226}=0, $R_{\varrho 237}$=$R_{\varrho 230}$=$R_{\varrho 226}$=1, R_{p234}=50, R_{p230}=5,000, R_{p229}=500 D_{p234}=D_{p230}=D_{p226}==0. 01m²/yr)

matrix diffusion and chemical sorption for multiple decay chain. In practice a numerical model for a three-member decay chain may suit better than a analytical model because the solutions of the latter for a three-member decay chain are expressed as multiple integral forms. But the physical meaning for matrix diffusion and chemical sorption during through a single fracture can be easily understood by the analysis of each physical term from the analytical solutions for a 3-member decay chain.

Np^{237}-U^{233}-Th^{229} and U^{234}-Th^{230}-Ra^{226} chain are selected among the multiple decay in the radioactive waste because Np^{237} and U^{234} have a considerable initial inventory in the waste solid or in the spent fuels.

The examples show that a daughter product will advance further along the fracture than its mother species of greater retardation factor though the half lives of the daughter are shorter. The concetration of radionuclides such as Th^{229} and Ra^{226} are greater than those of the mother at far field from the source though there is very small initial inventory in the waste solid or spent fuels.

The concentratons for a 3-member decay chain are compared with those of exact solution of approximation by neglecting radioactive decay [chamber, 1979] in the rock matrix. If the retardation factor of the mother is smaller than that of the daughter, the approximation will be incorrect. Be-

cause a precursor which decays to the daughter in the rock matrix is transported fast, the diffusion flux of the daughter at the surface in the rock matrix becomes smaller than that of radioactive decay in the rock matrix. But though the retardation factor of the mother is greater than that of the daughter, the approximation will be correct because the concentration of a precursor to decay to the daughter in the rock matrix is small.

The concentrations along the porous matrix decrease faster than those along the fracture. If the retardation factors of nuclides for a porous matrix are small, the variation of concentration along the porous matrix will become as important as those along the fracture.

References

1. J.H. Ahn, "Mass Transfer and Transport of Radionuclides in Fractured Porous Rock." p 1—10, 1988.

2. E.A Sudicky and E.O. Frind, "Contaminant Transport in Fractured Porous Media : Analytical Solution for a Two-Member Decay Chain in a Single Fracture." Water Resources Res., Vol. 20, No. 7, p1021—1029, July 1984.

3. Waste Isolation Systems Panel Board on Radioactive Waste Management Commisson on Physical Sciences, Mathmatics, and Resources National Research Council, "A Study of The Isolation System for Geologic Disposal of Radioactive

Wastes." National Academy Press Washington, C., p119—128, 1983.

4. J.H. Ahn, "Mass Transfer and Transport of Radionuclides in Fractured Porous Rock." p89—99, 1988.

5. P.D. Ritiger and N.J. Rose, "Differential Equations with Applications." Mcgraw-Hill, p267-303, 1985.

6. M.R. Spiegel, "Mathematical Handbook of Formulas and Tables." Mcgraw-Hill, p161—170, 1968.

7. 유영우, "단일균열 다공성암반에서 방사성핵종의 수송에 대한 3단계 붕괴사슬의 해석해," 석사학위논문, 1991. 8.

8. G.E. Grisak and J.F. Pickens, "Solute Transport Through Fractured Porous Media. 1. The Effect of Matrix Diffusion." Water Resources Res. Vol. 16, No. 4, p719—730, 1980.

9. D.H. Tang., E.O. Frind and E.A Sudicky, "Contaminant Transport in Fractured Porous : Analytical Solution for a Single Fracture." Water Resources Res., Vol. 17, No. 3, p555—565, 1981.

10. E.A Sudicky and E.O. Frind, "Contaminant Transport in Fractured Porous Media : Analytical Solution for a System of Parallel Fractures." Water Resoures Res., Vol. 18, No. 6, p1634—1642, 1982.

11. W. Ledermann, "Handbook of Applicable Mathmatics." Vol. 3, A Wiley-Interscience Pub., p540—542, 1981.