Journal of the Korean Nuclear Society
Volume 25, Number 4, December 1993

Continuous Time Markov Process Model for Nuclide Decay Chain Transport in the Fractured Rock Medium

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(Received September 6, 1993)

균열 암반 매질에서의 핵종의 붕괴사슬 이동을 위한 연속시간 마코프 프로세스 모델

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(1993. 9. 6 접수)

Abstract

A stochastic approach using continuous time Markov process is presented to model the one-dimensional nuclide transport in fractured rock media as a further extension for previous works [1-3]. Nuclide transport of decay chain of arbitrary length in the single planar fractured rock media in the vicinity of the radioactive waste repository is modeled using a continuous time Markov process. While most of analytical solutions for nuclide transport of decay chain deal with the limited length of decay chain, do not consider the case of having rock matrix diffusion, and have very complicated solution form, the present model offers rather a simplified solution in the form of expectance and its variance resulted from a stochastic modeling. As another deterministic way, even numerical models of decay chain transport, in most cases, show very complicated procedure to get the solution and large discrepancy for the exact solution as opposed to the stochastic model developed in this study. To demonstrate the use of the present model and to verify the model by comparing with the deterministic model, a specific illustration was made for the transport of a chain of three member in single fractured rock medium with constant groundwater flow rate in the fracture, which ignores the rock matrix diffusion and shows good capability to model the fractured media around the repository.

요 약

이전에 제시한 모델 1-3을 다시 확장하여 균열 암반에서의 일차원적 핵종이동에 관한 추 계적인 모델을 제시하였다. 이 모델은 처분장 근처의 암반내의 균열을 통한 무한 길이를 갖는 핵종의 붕괴

사슬에 의한 이동을 연속시간 마코프 프로세 스를 이용하여 모사한다. 이전의 결정론적 해석해에 의한 모델들이 균일한 다공성매질과 같은 단순성을 요구하고 핵종의 붕괴사슬의 수를 제한하며 균열암반매질내에서의 이동의 경우에는 균열에서 암반으로의 확산등이 고려되지 않거나 그 해의 형태가 복잡하다. 또다른 결정론적인 해를 제시하는 수치모델의 경우에도 해를 얻기 위한 과정이 상당히 복잡하 고 정확한 해를 제공하지는 못한다. 이에 반해 이 모델은 매질에서의 핵종의 농도에 관한 기대값과 그 분산으로서 비교적 용이하게 해를 제시한다. 모델을 검증하고 그 효율성과 정확성을 예시하기 위하여 암반으로의 확산이 무시된 단순화 된 매질에 대하여 3개의 붕괴사슬을 갖는 가상의 핵종에 대하여 이동거리와 시간에 대한 농도에 대하여 정확 한 해석해와의 비교가 행하여 졌다. 매질을 나눈 구획의 수에 종속 하는 수치분산을 보정하여 계산된 결과에서 이 모델이 해석해와 잘 일치하는 것을 알 수 있었다.

1. Introduction

In case of simulating nuclide transport through the geologic media around the repository in which radioactive wastes are disposed of, there have been various approaches either in deterministic way or in stochastic way.

Radionuclide transport in natural geologic media under variable field conditions has been found to be poorly described by the conventional deterministic advection-dispersion equations. Furthermore, even the information for the spatial variability of such system is comparatively well prepared, in most cases, there are many difficulties in formulating the deterministic models. To overcome these difficulties in modeling, stochastic approach has been used increasingly in the last few years. In a stochastic approach, parameters involved in the transport in geologic system are regarded as random variables characterized by probability distributions rather than by well-defined deterministic values.

A stochastic approach by which the concentration distribution as a result of nuclide transport of decay chain of arbitrary length in heterogeneous media such as fractured rock media could be modeled is proposed using a continuous time Markov process. In three previous papers, Lee et al. [1-3] have already successfully used a continuous time Markov process to model the nuclide distributions in geological systems. The first paper dealt with one dimensional nuclide transport in homogeneous porous media whereas the rest two papers discussed nuclide transport in fractured rock media where a special process of diffusive loss into the rock matrix from the fracture is considered. In the fractured rock medium groundwater flow occurs mainly within the planar fracture because of the low permeability usually associated with the rock matrix. Thus the fracture offers main pathway for nuclide transport rather than the rock matrix. Because the process related to the diffusion into the rock matrix from the fracture varies with time, the application of continuous time Markov process model for the fractured rock medium is no more time-homogeneous unlike the former case.

In using a continuous time Markov process, the media can be considered as a series of discretized compartments and the nuclide concentration in each compartment can be considered as a time-dependent random variable.

A nuclide in a given time interval could make a transit to any compartment by groundwater flow, could form as decay products from its parent nuclide or could also disappear from any present compartment due to radioactive decay or diffusive loss. All these processes are obviously conditional only on the present location of the nuclide regardless of its previous history utilizing the Markov conceptualization of the geologic system.

As mentioned earlier, in general, nuclide transport will be dominated along the fracture. Although this is true, the rock matrix adjacent to the fracture plays an important role in overall nuclide transport. A convenient way to study such transport is to consider the rock matrix has a single planar fracture as discussed by many authors. [4,5] However, many of these models are not only on the deterministic base but limited to single nuclide transport.

There are many works related to the chain nuclide transport in both forms of analytical and numerical models. [6,7] As for the analytical models, however, these studies almost consider only homogeneous porous media. Among numerical models for the fractured rock media, even large part of these model still have the same situation, several models for the fractured rock media considering the rock matrix system are available. [e.g. 8]

Even though an analytical solution of two decay chain transport has been developed by Sudicky et al. [9] for the fractured rock media, this is limited by the length of decay chain and has a very complicated solution form.

This paper, as another extended approach of previous works, deals with a stochastic model for the nuclide transport of decay chain of arbitrary length in the fractured rock media in the vicinity of the radioactive waste repository and also offers the mean values and variance of the state variables, as the primary desired quantities from a stochastic model here the nuclide concentration in the fractured rock media, as a function of time.

2. Nuclide Distributions in the Fracture

A continuous-time Markov process $\{X(t),\ t\geq 0\}$ is a stochastic process having the property that the conditional distribution of the future state j at time $t+s$, given the present state i at time s and all past states, depends only on the present state i and is independent of the past.

Utilizing this Markov conceptualization, in matrix notation, the relation between the rate of change of the transition probability and the intensity of transition is represented as [1]

$$\frac{d}{dt}\mathbf{P}(t)=\mathbf{P}(t)\,\mathbf{\Lambda}(t) \tag{1}$$

which is called Kolmogorov forward differential equations and

$$\mathbf{P}(0)=\mathbf{I}(\text{the identity matrix}) \tag{2}$$

where

$\mathbf{P}(t)$ = transition probability matrix, $P_{ij}(t)$.

A serial compartment i of rock matrix system in groundwater-saturated porous rock of porosity ϕ_{pi} ($i=1,\ 2,\ \cdots,\ N$) containing a single planar fracture of half width b^i is considered. The fracture can be considered as a finite number of N compartments within which complete mixing of nuclides with groundwater takes place instantly. As considered by Tang et al. [5], the permeability of the porous matrix is very low and then transport is dominated in the fracture. In the rock matrix nuclide will be transported by molecular diffusion in the direction perpendicular to the direction of the axis of the fracture. A decaying nuclide source locates at the inlet of the fracture.

The following processes are to be considered probabilistically to obtain the nuclide distribution in the fracture : (1) transition by the groundwater flow, (2) molecular diffusion from the fracture into the rock matrix, (3) adsorption onto wall of the fracture and within the rock matrix, and (4) radioactive decay chain transport through the fractured rock media is preassumed. Longitudinal dispersion in the fracture, however, is assumed to be negligible in this study.

Once such a geologic system is assumed to have Markov property, since the Markov process requires that only the present value of the time dependent random variable (i. e., time-dependent number of nuclides or concentration in certain compartment) be known to determine the future value of the random variable, the nuclide transport in geologic media, which is divided by finite number of geologic compartments N, can be modeled as a time continuous Markov process, which is continuous in time with respect to the individual transport processes but discrete in medium.

At any time $\tau \in [0, t)$, when nuclide component $l(l=1, 2, \cdots)$ adds to the first compartment at the rate of $\xi(\tau)$, that is equal to the volumetric flow rate of nuclides into the first compartment and may be represented as

$$\zeta^l(\tau) = Q_{in} C_0^l(\tau)\ V, \qquad l = 1, 2, \cdots \tag{3}$$

where

Q_{in}=volumetric flow rate of feeding groundwater into the first compartment ($L^3\ T^{-1}$)

V=volume of the first compartment (L^3).

$C_0^l(\tau)$=source concentration of the lth member at time τ (L^{-3}), which is given by the solution of Bateman's differential equations as, with l ranging from 1 to m

$$C_0^l(\tau) = \tilde{C}_0^l e^{-\lambda^l t} + \lambda^{l-1}\tilde{C}_0^{l-1} \sum_{m=l-1}^{l} \frac{e^{-\lambda^m t}}{\prod_{\substack{k=l-1\\ k\neq m}}^{l} (\lambda^k - \lambda^m)}$$

$$+ \cdots + \lambda^{l-1}\lambda^{l-2}\cdots\lambda^1 \tilde{C}_0^1 \sum_{m=1}^{l} \frac{e^{-\lambda^m t}}{\prod_{\substack{k=1\\ k\neq m}}^{l} (\lambda^k - \lambda^m)}, \qquad l = 1, 2, \cdots \tag{4}$$

where

$\tilde{C}_0^l$=source concentration of the lth member at time 0 (L^{-3}).

λ^l=decay constant of the lth member (T^{-1}).

Similarly, by analogy, at any time $\tau \in [0, t)$, the change rate of nuclides into daughter component from its parent nuclide, $\xi_i^l(\tau)$ is represented by

$$\xi_i^l(\tau) = \lambda^l C_i^l(\tau)\ V_i,\ i = 1, 2, \cdots, N,\ l = 1, 2, \cdots \tag{5}$$

where

$C_i^l(\tau)$=concentration of lth member at time τ (L^{-3})

As soon as a nuclide which fed freshly by supplying at the inlet of the system (Equation (3)) or by forming due to the radioactive decay of its precursor enters the system (Equation (5)), it may begin to transfer to one of the other compartments at once or may disappear. Here we can assume that all nuclides in the system behave stochastically and independently one another.

The number of new nuclides that enter the first compartment from source nuclides during time interval $d\tau$ is $\zeta^l(\tau)\ d\tau$. If we consider the number of nuclides that have successfully entered the first compartment, then it has a respective probability of $P_{1j}(t-\tau)$ that nuclides exist in j at time t, i.e., transit from the first compartment to compartment j during the time interval $(t-\tau)$. Therefore a binomial distribution can be formed for these new nuclides.

Also for large value of $\zeta^l(\tau)$ dt, the binomial distribution is approximated to Poisson distribution.

In similar way the probability that $\xi_i^l(\tau)\ d\tau$, which is the number of new nuclides that form in the ith compartment from the precursor nuclides in time interval $d\tau$ will exist in j at time t during the time interval $(t-\tau)$ can be expressed as $P_{ij}(t-\tau)$. The probability distribution may also be considered as Poisson distribution.

Then we can get $X_j^l(t)$, $E[X_j^l(t)]$ and its variance Var $[X_j^l(t)]$ at time t as defined in previous paper [1-3] :

$$E\left[X_j^l(t)\right] = \sum_{i=1}^{N} m_i^l(0) P_{ij}(t) + \int_0^t$$

$$\left\{\zeta^l(\tau)P_{1j}(t-\tau)+\sum_{i=1}^{N}\xi_i^l(\tau)P_{ij}(t-\tau)\right\}d\tau,$$

$$j=1,2,\cdots,N,\ l=1,2,\cdots \quad (6)$$

$$\mathrm{Var}\left[X_j^l(t)\right]=\sum_{i=1}^{N}m_i^l(0)P_{ij}(t)\{1-P_{ij}(t)\}+\int_0^t$$

$$\left\{\zeta^l(\tau)P_{1j}(t-\tau)+\sum_{i=1}^{N}\xi_i^l(\tau)P_{ij}(t-\tau)\right\}d\tau,$$

$$j=1,2,\cdots,l=1,2,\cdots \quad (7)$$

The first term of R. H. S. of the Equation (6) represents the number of nuclides in the ith compartment initially at time zero which have been survived and entered *j*th compartment to time t. Two terms of integrand represent the number of nuclides survived during the time interval [0, t) and originally due to feed of new nuclides and decay of parent nuclides, respectively.

Therefore, the expected value and variance of $C_j^l(t)$, concentration of nuclides in j at time t can be expressed, respectively, as

$$\mathrm{E}\left[C_j^l(t)\right]=\frac{\mathrm{E}\left[X_j^l(t)\right]}{V_j},j=1,2,\cdots,N,\ l=1,2,\cdots \quad (8)$$

$$\mathrm{Var}\left[C_j^l(t)\right]=\frac{\mathrm{Var}\left[X_j^l(t)\right]}{V_j},j=1,2,\cdots,N,\ l=1,2,\cdots \quad (9)$$

where

V_j=pore water volume of compartment j of the fracture (L^3).

The transition probability from a compartment i to another compartment j is affected by the intensity of transition. This intensity of transition is related to the process involved. The diffusive transport of nuclide which is assumed to be negligible compared to advective transport for the media having large Peclet number and another diffusive loss term into the rock matrix are also excluded in this case.

For simplicity and also reasonably, the groundwater flow and nuclide transport are assumed to be made only between adjacent compartments.

The intensity of transition $h_{ij}(T^{-1})$ for the groundwater flow through some pore volume in porous medium can be written as

$$h_{ij}=\frac{Q_{ij}}{V_i} \quad (10)$$

where

Q_{ij}=volumetric flow rate from compartment i to compartment j ($L^3\ T^{-1}$)

V_i=volume of compartment i (L^3).

Assuming that flow is well mixed instantly with regard to groundwater and nuclides, the transition probability due to advection can be written as [e. g. 10]

$h_{ij}\Delta t+o(\Delta t)$=Pr {a nuclide in i at time t will be in j at time($t+\Delta t$)} (11)

Similarly nuclide may decay out from compartment i at a rate represented by decay constant λ^l (T^{-1}). Therefore,

$\lambda_l\Delta t+o(\Delta t)$=Pr {a nuclide in i at time t will be decayed out at time($t+\Delta t$)} (12)

Under the assumption of linear isotherm sorption of nuclides in the compartment i, h_{ij} can be replaced by h_{ij}/R_i^l, where R_i^l is retardation coefficient in the ith compartment of the fracture. Then

$$\lambda_{ij}^l=\sum_{j\neq i}\frac{h_{ij}}{R_i^l},\ i=1,2,\cdots,N,\ l=1,2,\cdots \quad (13)$$

With these the probability that the nuclide will remain at time $t+\Delta t$ in i without making any transition or disappearance is $\{1-(\Sigma_{j=l}\lambda_{ii}^l\Delta t+\lambda^l\Delta t)+o(t)\}$, from which, if this probability is denoted by $\{1+\lambda_{ii}^l\Delta t+o(\Delta t)\}$ in case of no diffusive loss into the rock matrix,

$$\lambda_{ii}^{l} = -\left[\sum_{j \neq i} \frac{h_{ij}}{R_i^l} + \lambda^l\right], \; i = 1, 2, \cdots, N, \; l = 1, 2, \cdots \quad (14)$$

where λ_{ii}^{l} is interpreted as the negative sum of all probabilities of exit from compartment i.

When the diffusive loss into the rock matrix in the direction perpendicular to the fracture from the fracture is considered, the corresponding intensity of transition for diffusive loss $\lambda_{diffi}^{l}(t)$ (T^{-1}) into rock matrix can be expressed as

$$\lambda_{diffi}^{l}(t) = \frac{\phi_{pi}}{b_i}\left\{\sqrt{\frac{D_{pi}}{\pi R_{pi}^{l}\, t}}\right.$$

$$\left.\exp\left(-\frac{b^2 R_{pi}^{l}}{4 D_{pi}^{l}\, t}\right)\right\}, \; i = 1, 2, \cdots, N, \; l = 1, 2, \cdots \quad (15)$$

Equations (13), (14) and (15) can be integrated, under the assumption that the transport is considered to be made only between adjacent compartments, into the transition coefficient matrix as follows :

$$\left(\lambda_{ij}^{l}(t)\right) = \begin{cases} -\left[\frac{h_{ij}}{R_{fi}^{l}} + \lambda^l + \lambda_{diffi}^{l}(t)\right], & j = i \\ \frac{h_{ij}}{R_i^{l}}, & j = i+1 \\ 0, & \text{otherwise} \end{cases},$$

$$i = 1, 2, \cdots, N, \; l = 1, 2, \cdots \quad (16)$$

Meanwhile, in order to evaluate Equations (1) and (6) having inhomogeneous intensity of transition, brief algorithm for the discrete time approximation scheme represented in previous papers [2,3] can be discribed as follows.

Rewiring the Equation (6) compactly as, abbreviating the superscripts,

$$E[X_j(t)] = \sum_{i=1}^{N} m_i(0) P_{ij}(t)$$

$$+ \int_0^t \sum_{i=1}^{N} \omega_i(\tau) P_{ij}(t-\tau) \; d\tau, \; j = 1, 2, \cdots, N \quad (17)$$

where

$$\omega_i(\tau) = \begin{cases} \zeta_i(\tau) + \xi_i(\tau), & i = 1 \\ \xi_i(\tau), & i = 2, 3, \cdots, N \end{cases} \quad (18)$$

If $\lambda_{ji}^{i}(t)$ and $\omega_i(t)$ is piecewise constant (i. e., $-_{ij}^{l}$ (t)$= \lambda_{ij}^{l}(T)$ and $\omega_i(\tau) = \omega_i(T)$) during short time interval [0, T<t), then the solution of the Equation (17) may be written as

$$E[X(T)] = e^{\Lambda T} E[X(0)] + \int_0^T e^{\Lambda T} \, d\tau \, \Omega(T) \quad (19)$$

where **X** and **W** are matrix forms of X_j and ω_i, respectively and $e^{\Lambda T}$, the solution of the Equation (1) is given, subject to the initial condition **P**(0)=**I**, where **I** is the identity matrix, as

$$P(T) = e^{\Lambda T} P(0) = e^{\Lambda T} \quad (20)$$

Similarly, during time interval $[T, 2T<t)$ Equation (19) becomes under the assumption of constant $\lambda_{ij}^{l}(2T)$ and $\omega_i(2T)$

$$E[X(2T)] = e^{\Lambda T} E[X(T)] + \int_T^{2T} e^{\Lambda(2T-\tau)} \, d\tau \, \Omega(2T) \quad (21)$$

Now, in general, after ($N-1$)th step, for the time interval of $[nT, (n+1)T)$,

$$P[(N+1)T] = e^{\Lambda T} P(NT) = e^{(N+1)T} \quad (22)$$

and accordingly

$$E[X(N+1)T] = e^{\Lambda(N+1)T} E[X(NT)] + \int_{NT}^{(N+1)T} e^{\Lambda[(N+1)T-\tau]} \, d\tau \, \Omega(NT)$$

$$= e^{\Lambda(N+1)T} E[X(NT)] + \int_0^T e^{\Lambda \tau} \, d\tau \, \Omega(NT) \quad (22)$$

Therefore, we need only evaluate the matrix exponential of the transition probability and its integration as seen in the R. H. S. of the above equation.

3. Numerical Illustration

To demonstrate the use of the present model and to verify the model by comparing with the deterministic model, a specific illustration was made for the transport of a chain of three members in single-fractured rock medium with constant groundwater flow rate in the fracture.

For simplicity in comparison with available analytical solution, we introduce a computational result only for the case of no diffusion into the rock matrix, which yields one dimensional model for porous medium. The effects of the diffusive loss into the rock matrix can be incorporated by a straightforward extension of the present illustration ; however, in order to carry out direct comparison with the analytical solution for the equivalent system it hase been ommitted. And such assumption does not affect on the verification of the whole model.

We further assumed the retardation coefficients of the lth member in the whole medium are equal, i. e., $R_i^l = R^l$ for all l.

The properties of three members and medium are given in Table 1. For the purpose of comparison, some of these data were chosen arbitrarily or to correspond to the data used by Huyakorn et al. [8]

Initial concentration values of the second and third member equal to 0.0 implies that all of the source concentration originally prepared in the inlet of the fracture is in the form of the first member of the chain. The parent member has a initial concentration of unity.

A result by our Markov model was compared against those obtained by Lung[7] through his code UCB-NE-40, designed for general solution for a chain of arbitrary length through the semi-infinite homogeneous medium.

In Figure 1, the concentration profiles of the first, second, and third member, normalized to initial concentration of the first member at time 0 as a function of distance at times equal to 100 and 1000 years, respectively, by the present model for two different cases are plotted showing good comparison of corresponding analytical solution : one is for the retardation of 9532 at the time of 1000 years and the other for the retardation of 100 at the time of 100 years. These retardations and time values are arbitrarily chosen. It is easily seen that in both cases with and without retardation and radioactive decay, there exists excellent agreement between the results using the present model and the analytical solutions. Even though the retardation coefficients are the same for all members the profiles at different times indicate that the third member of the deacy chain can travel further along the fracture than the first and second since the half-life of the third member is approximately 15 and 436 times longer than those of the first and second, respectively. Then it is to be expected that the profiles of $C^{(1)}$ and $C^{(2)}$ at each time will tend to lag behind those of $C^{(3)}$.

Breakthrough curves of the three members are also plotted showing comparison of corresponding analytical solution in Figure 2 as a function of time at distances of 20 and 50 m, respectively. Parameter values are the same to those used for Figure 1.

Table 1. Input Parameters

Chain l	(1)→(2)→(3)
$C_0^{(1)}$	1.0
$C_0^{(2)}$	0.0
$C_0^{(3)}$	0.0
$R^1(^{1)}$	9532.0(100.0)
$R^1(^{2)}$	9532.0(100.0)
$R^1(^{3)}$	9532.0(100.0)
L(m)(along the fracture)	100
$\lambda^{(1)}$(1/yr)	0.0016
$\lambda^{(2)}$(1/yr)	0.04620
$\lambda^{(3)}$(1/yr)	0.000106
q/ϕ (m/yr)	10.0
D(m^2/yr)(for UCB-NE-40)	25.0[a]
N(for Markov model)	20

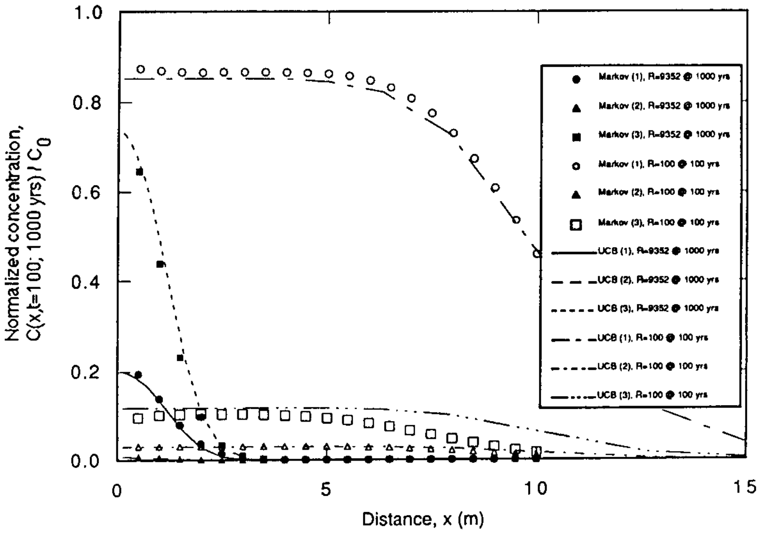

Fig. 1. Concentration Profiles of the First, Second, and Third Components by Continuous-time Markov Model, Showing Comparison of Corresponding Analytical Solutions.(Chain.grf)

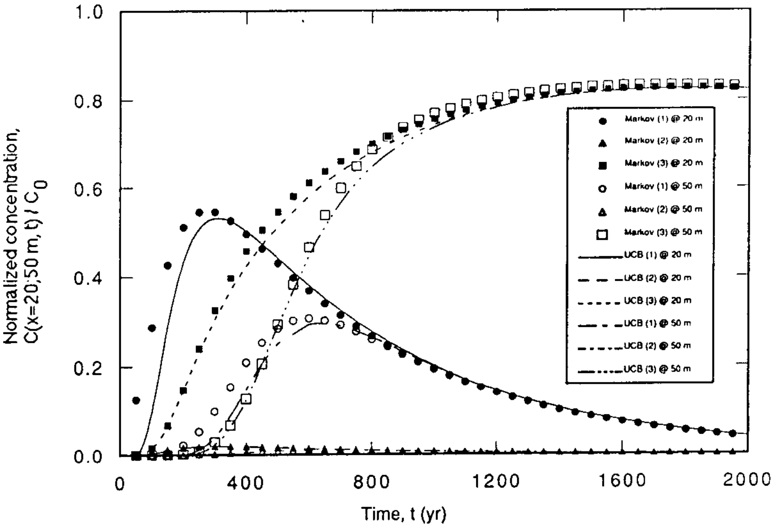

Fig. 2. Breakthrough Curves of the First, Second, and Third Components, Showing Comparison of Analytical Solutions for Two Different Distances.(Chainb.grf)

4. Conclusion

Through this study a stochastic modeling using a continuous time Markov process has been made as a further extended approach of two previous works. It deals with the nuclide transport of decay chain of arbitrary length in the fractured rock media in the vicinity of the radioactive waste repository.

Since this model is discrete in medium space, physical and geochemical parameters including groundwater velocity, dispersion coefficient, retardation coefficients, and losses due to radioactive decay or diffusion out of the system, which affect nuclide transport, can be easily incorporated for such heterogeneous media as fractured rock medium having spatially varied parameters.

The Markov process model developed in this study, with small compensating of dispersion coefficient by the same way to that represented in the previous paper [1–3], which are known to be sensitive to the number of discretized compartment showing numerical dispersion as the number of compartments is increased, the model agrees well to analytical solution.

It was illustrated that a simplified example compared to an analytical solution, which ignores the rock matrix diffusion shows good capability to model the fractured media around the repository.

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