

# A Theoretical Approach on the Migration of a Chelating Radionuclide in Porous Medium

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## ABSTRACT

A new model was developed in order to investigate the effects of chelating agents on the migration of a radionuclide in the form of ion or chelate. The migration behavior of the chelated radionuclide was analyzed by formulating a convective-dispersion transport equation which included a degradation of chelating agent and chelated radionuclide. The mathematical model was analytically solved and checked with the existing retardation factor. The results show that the migration velocity of the chelated radionuclide was much faster than the ionic one due to the decreased retardation. Therefore, it was concluded that a new remedial action should be developed to reduce the generation and release of chelating agents from the nuclear power plant into the environment.

*Key Words : Radionuclide migration, Chelated radionuclide, Adsorption, Degradation*

## INTRODUCTION

A wide range of chemical and physico-chemical reactions can be considered to influence the migration phenomena of radionuclides of various forms in geological radioactive waste disposal sites. Possible reactions may be radioactive decay, sorption, precipitation, dissolution, filtration, degradations, hydrolysis, complexation, and colloid formation. However, an urgent need for information has led to research on relatively limited aspect of these complex problem and a tendency to oversimplify the simulation of natural conditions.

It has been within only the past decade that the important effect of organic substances on the environmental geochemistry of trace metals have truly been recognized. Also it has been stated [1] that complex formation of radionuclides with complexing agents found in ground water significantly influenced the migration behavior of the radionuclides. Because it forms extremely strong complexes with rare earths and actinides, ethylenediaminetetraacetic acid (EDTA) or similar chelating agents may also be contributing to the mobilization of these radionuclides from various terrestrial radioactive waste burial sites. For

example, they have been measured that although the distribution coefficient,  $K_d$  values for  $^{60}\text{Co}$  in weathered Conasauga shale at pH 6.7 and 12 were approximately  $7.0 \times 10^4$  and  $10.12 \times 10^4 \text{ ml/g}$ , respectively, the  $K_d$  values were reduced to 2.9 and 0.8 ml/g in the presence of  $10^{-5}$  MEDTA [1].

The organic materials can be found in ground water in radioactive waste repository (especially in low level radioactive waste repository) such two categories as natural (e. g., humic or fulvic substances) and synthetic (e. g., decontamination agents such as EDTA, citric acid and picolinic acid) organics [2]. The most abundant and important natural organic material in humic substances, which is consisted of humic acid and fulvic acid [3]. One important property of humic substances is their ability to form complexes with a variety of polyvalent metals. For example, humic substances may cause reduction of iron(III) to iron(II) and subsequent dissolution and depolymerisation of colloidal and particulate iron. Synthetic organic compounds such as EDTA, which is used in nuclear decontamination solutions, increase radionuclide transport rates in at least two different low-level waste burial ground [1, 4]. Means et al. [1] identified the importance of synthetic chelating agents in the transport of  $^{60}\text{Co}$  in low-level repositories. Cleveland and Rees [4] discovered similar migration Pu-chelate complexes in Maxey Flat trench leachates.

In order to analytically predict the transport of radionuclides in a porous medium which is affected by the presence of complexing agents, it is necessary to develop a complete mathematical model. This means that the mechanisms must be described and the governing equation derived, along with their general solutions for the transport

processes. In this study, the five mechanisms of advection, dispersion, adsorption, chelation, and degradation are described and mathematically modelled. A general convection-dispersion transport equation is used to predict the migration of radionuclides in the presence of chelating agents in porous media.

## TRANSPORT MECHANISMS

### Adsorption

The adsorption process can be characterized by isotherms that describe the equilibrium relationship between adsorbed and aqueous contaminant concentrations at a given temperature. The adsorption isotherm of a given contaminant and soil material can be estimated by several experimental techniques, including measuring the relationship between the adsorbed and aqueous concentrations of a given contaminant by the batch equilibrium technique. This method has the advantage of directly establishing the linearity of the isotherm and, if a desorption isotherm is measured, the reversibility of the sorption process.

If the isotherm is linear and reversible, the isotherm is uniquely defined by a distribution coefficient  $K_c$ , which is constant and equal to the slope of the isotherm. Several investigators [6, 7] have measured linear isotherms for adsorption of nonionic organic solutes onto soils, sediments, and aquifer materials. These isotherms may be described by:

$$S = K_d \cdot C \quad (1)$$

where

S = contaminant concentration in the adsorbed solid phase [mole/kg],

$C$  = contaminant concentration in the solution phase [mole/m<sup>3</sup>],

$K_d$  = distribution coefficient [m<sup>3</sup>/kg].

Results from such batch equilibrium adsorption studies may be used for estimating the mobility of metal ions and metal chelates in ground water. The migration of a sorbing contaminant in ground water is retarded relative to the movement of a noninteracting tracer by a factor known as the retardation factor  $R$ . The retardation factor, accompanied by the linear isotherm presented by distribution coefficient of Eq.(1), can be obtained as follows:

$$R = 1 + \frac{(1-\theta)\rho_b}{\theta} K_d \quad (2)$$

where  $\rho_b$  is the bulk density of soil material [kg/m<sup>3</sup>].

### Chelation

For the expression of chelate formation, we introduce the conditional stability constant as an equilibrium constant for the chelate formation reaction. Ringbom [8] has used this to calculate the influence of a whole series of side reaction other than main complex formation reaction:

- 1) hydrolytic reactions of the complexing agents and of the metal ion,
- 2) formation of acid or base complexes,
- 3) competing side reactions of the complexing agent with other metal ions presents, and
- 4) reactions of metal ions with buffering and masking substances.

He introduced the term "conditional constant" in order to stress that the constant is not constant

but depends on the experimental conditions.

The general form of the overall conditional stability constant for a mononuclear complex (i. e., chelate) is given as follows [9]:

$$\beta'_n = \frac{[ML_n]}{[M][L]^n} \quad (3)$$

where  $[M]$  is the apparent free metal ion concentration [mole/m<sup>3</sup>], i. e., the concentration of radionuclide ion that has not chelated. Similarly,  $[L]$  is the apparent ligand concentration [mole/m<sup>3</sup>], i. e., the concentration of the ligand not bound to the central ion  $M$ , whether  $L$  is in protonated or non-protonated form, or in the form of complexes with other radionuclide ions. And  $n$  is the average number of ligands per radionuclide ion.

Typically the ligand concentration will be significantly greater than the radionuclide ions. Hence the ligand concentration  $[L]$  can be approximated by the initial loading ligand concentration  $[L_0]$ . Using the conditional stability constant concept we can find the relationships between the concentrations of chelated and ionic radionuclides given as follows:

$$\frac{[ML_n]}{[M]} = \beta'_n [L_0]^n = K_M \quad (4)$$

where  $K_M$  is the apparent stability constant of the chelate [dimensionless].

### Degradation

Degradation process of the migrating contaminant includes all the irreversible physical, chemical, and biological processes. Also the chelated

radionuclides have complex migration processes such as filtration, chemical transformation, precipitation, and decomposition. These processes may affect the migration of contaminant in porous medium. The often mentioned models affecting the migration of contaminant are radioactive decay, precipitation, hydrolysis, oxidation-reduction, microbial transformation, and chelating agents degradation. All the processes except for the radioactive decay are complex functions of the environmental and biological parameters. Degradation of chelating agents are important factor in predicting the fate of a chelating agents in the environment. The degradation of chelating agents includes several types of degradation mechanisms such as biological photolytic, radiolytic, chemical, and thermal degradations. Detailed explanations are presented by other studies [10, 11, 12].

To model the degradation of the chelating agent, a first-order kinetic model is also used.

$$\frac{dL}{dt} = -k_{dc} L \quad (5)$$

where,

$L$  = concentration of the chelating agents  
[mole/m<sup>3</sup>],

$k_{dc}$  = degradation rate constant of the chelating agents [y<sup>-1</sup>].

Degradation rate constant  $k_{dc}$  possibly includes all kinds of degradation phenomena and may be dependent on temperature and environmental conditions. Since the chelating agent reacts and forms a strong chelate with a radionuclide ion, the same process of the degradation with chelating agent may be also applicable to the degradation of a

chelate. Therefore, in this study, a first-order kinetic model is also used for the degradation reactions of a chelated radionuclide considered.

## ANALYSIS

### Governing equation

Consider a radioactive solute as a free metal ion at concentration in pore water in porous soil medium. Also present in porous medium is chelating agent, on which an ionic radionuclide can form chelate with the chelating agent. We assume the one-dimensional convection-diffusive transport within the porous medium for the ionic radionuclide and chelated radionuclide. A number of assumptions are made as follows:

- 1) Initially, a radionuclides is dissolved as an ionic and a chelated form in the groundwater in a porous medium.
- 2) The groundwater flows through the medium at a constant rate, in one direction.
- 3) The groundwater and the medium are incompressible.
- 4) The medium is saturated, isotropic, and homogeneous. Hence interaction between the medium and the dissolved radionuclide is the same throughout the considered system, and the dispersivity is also constant.

The equation governing the transport of ionic radionuclide is given as follows:

$$\frac{\partial}{\partial t} \left( C_1 + \frac{(1-\theta)\rho_b}{\theta} S_1 \right) = D_1 \frac{\partial^2 C_1}{\partial x^2} - V_1 \frac{\partial C_1}{\partial x} - \lambda_1 \left( C_1 + \frac{(1-\theta)\rho_b}{\theta} S_1 \right) \quad (6)$$

$$x > 0, t > 0$$

where

$C_1$  = concentration of an ionic radionuclide in the pore water solution phase [mole/ $m^3$ ],

$S_1$  = concentration of an ionic radionuclide in the solid phase [mole/kg],

$D_1$  = dispersion coefficient of ionic radionuclide in the porous medium [ $m^2/y$ ],

$V_1$  = average pore water velocity in the porous medium [ $m/y$ ],

$\lambda_1$  = decay constant of ionic radionuclide [ $y^{-1}$ ],

$x$  = distance [ $m$ ],

$t$  = time [ $y$ ].

For the same species as a chelate, the governing equation is given as follows:

$$\frac{\partial}{\partial t} \left[ C_2 + \frac{(1-\theta)\rho_b}{\theta} S_2 \right] = D_2 \frac{\partial^2 C_2}{\partial x^2} - V_2 \frac{\partial C_2}{\partial x} - (\lambda_2 + k_{dc}) \left[ C_2 + \frac{(1-\theta)\rho_b}{\theta} S_2 \right] \quad (7)$$

$$x > 0, t > 0$$

where

$C_2$  = concentration of a chelated radionuclide in the solution phase [mole/ $m^3$ ],

$S_2$  = concentration of a chelated radionuclide in the solid phase [mole/kg],

$D_2$  = dispersion coefficient of chelated radionuclide in the porous medium [ $m^2/y$ ],

$V_2$  = average pore water velocity in the porous medium [ $m/y$ ],

$\lambda_2$  = decay constant of the chelated radionuclide [ $y^{-1}$ ],

$k_{dc}$  = degradation coefficient of the chelated radionuclide in pore water [ $y^{-1}$ ].

We assume linear adsorption equilibrium between the ionic radionuclide species in liquid and

the same species adsorbed onto the solid. Also, the chelates both in liquid and solid are assumed to undergo linear adsorption equilibrium and given as follows:

$$K_{d1} = \frac{S_1}{C_1}, \quad K_{d2} = \frac{S_2}{C_2} \quad (8)$$

where

$K_{d1}$  = distribution coefficient of the ionic radionuclide [ $m^3/kg$ ],

$K_{d2}$  = distribution coefficient of the chelated radionuclide [ $m^3/kg$ ].

For the expression of the relationship between the concentration of the ionic radionuclide and chelated radionuclide in the solution (i. e., relationship of  $C_1$  and  $C_2$ ), Eq. (4) is used and  $K_M$  is assumed to be constant.

$$C_2 = \beta_n' [L_0]^n C_1 = K_M C_1 \quad (9)$$

where  $K_M$  means the mole ratio of the ionic radionuclide to the chelated radionuclide in the solution.

Adding Eqs. (6) and (7), we can obtain the equation for  $C_2$  in terms of the effective retardation factor  $R$ , effective dispersion coefficient  $D$ , effective velocity  $V$ , and effective total degradation constant  $K_{dc}$ . Then overall governing equation can be given as follows:

$$R \frac{\partial C_2}{\partial t} = D \frac{\partial^2 C_2}{\partial x^2} - V \frac{\partial C_2}{\partial x} - K_{dc} C_2 \quad (10)$$

where

$$R = \frac{R_1}{K_M} + R_2 \quad (11)$$

$$D = \frac{D_1}{K_M} + D_2 \quad (12)$$

$$V = \frac{(K_M + 1)}{K_M} V_1 \quad \text{since } V_1 = V_2 \quad (13)$$

$$K_{dc} = R \lambda_1 + R_2 k_{dc} \quad \text{since } \lambda_1 = \lambda_2 \quad (14)$$

In the above equations, the effective retardation factor  $R$ , effective dispersion coefficient  $D$ , effective velocity  $V$ , and effective total degradation constant  $K_{dc}$  are given as complex forms incorporating the term for the ionic radionuclide and the term for the chelated radionuclide.  $K_M$  acts as a weighting factor controlling the relative importance of one of two terms in the complexed parameters.

Similarly, for  $C_1$ , the major parameters are also given as follows:

$$R' = R_1 + K_M R_2 \quad (15)$$

$$D' = D_1 + K_M D_2 \quad (16)$$

$$V' = (1 + K_M) V_1 \quad \text{since } V_1 = V_2 \quad (17)$$

$$K'_{dc} = R \lambda_1 + R_2 K_M K_{dc} \quad \text{since } \lambda_1 = \lambda_2 \quad (18)$$

### Solution of the equation

The initial condition at  $t=0$  is given as follows:

$$C_1 = C_2 = 0 \quad \text{at } t=0, \text{ for } x \geq 0. \quad (19)$$

The boundary conditions based upon the assumption of semi-infinite medium is given as follows:

$$\frac{\partial C_1(x,t)}{\partial X} = \frac{\partial C_2(x,t)}{\partial x} = 0 \quad \text{as } x \rightarrow \infty, \text{ for } t > 0 \quad (20)$$

$$\left[ -D \frac{\partial C_1}{\partial X} + VC_1 \right] = VC_{10} \quad \text{at } x=0 \text{ for } t > 0 \quad (21a)$$

$$\left[ -D \frac{\partial C_2}{\partial X} + VC_2 \right] = V C_{20} \quad \text{at } x=0 \text{ for } t > 0 \quad (21b)$$

where  $C_{10}$  and  $C_{20}$  are the initial concentrations of ionic radionuclide [ $\text{mole/m}^3$ ] and chelated radionuclide [ $\text{mole/m}^3$ ] in the solution, respectively.

For a more detailed analysis of the source term conditions, two cases can be considered. In the absence of chelating agents, a dissolved radionuclide as only an ionic form are released at constant concentration  $C_s$  [ $\text{mole/m}^3$ ], i. e.,

$$C = C_s \quad \text{at } x=0, \text{ for } t > 0 \quad (22)$$

However, in the presence of chelating agents, the dissolved radionuclide is released as both of ionic and chelated forms at constant concentration  $C_0$  [ $\text{mole/m}^3$ ], i. e.,

$$C_0 = C_x + C_{20} = \left[ \frac{K_M + 1}{K_M} \right] C_{20} \quad (23)$$

The governing equation for  $C_2$ , Eq. (10), subject to the conditions Eqs. (20) and (21) is solved by the analytic method presented by van Genuchten et. al. [13] and given as follows:

$$\begin{aligned} \frac{C(x,t)}{C_0} = & \frac{V}{V+\beta} \exp\left\{\frac{x(V-\beta)}{2D}\right\} \text{efc}\left\{\frac{Rx-\beta t}{2(DRt)}\right\}^{1/2} \\ & + \frac{V}{V-\beta} \exp\left\{\frac{x(V+\beta)}{2D}\right\} \text{efc}\left\{\frac{Rx-\beta t}{2(DRt)}\right\}^{1/2} \\ & + \frac{V^2}{2DRt} \exp\left\{\frac{Vx}{D} - K_{dc}t\right\} \text{efc}\left\{\frac{Rx+Vt}{2(DRt)}\right\}^{1/2} \end{aligned} \quad (24)$$

where

$$\beta = \left[ V^2 + 4DRK_{dc} \right]^{1/2} \quad (25)$$

## RESULTS AND DISCUSSION

### Retardation factor

Retardation factor is one of the important parameters for predicting radionuclide migration in geologic repositories. Therefore, radionuclide retardation factors must be measured to provide this input parameter for the prediction. The retar-

dation factor is usually determined in the laboratory using a batch method based on the relationship between a retardation factor and a distribution coefficient presented in Eq. (2). A retardation factor has also been obtained by the column method using collected disturbed or undisturbed soil [14, 15]. Since the retardation factor is the ratio of water velocity to the migration speed of a radionuclide, whether it is ion or not, the retardation factor is defined as follows:

$$R_D \equiv \frac{V_w}{V_s} \quad (26)$$

where  $V_w$  is an average ground water velocity [m/y] and  $V_s$  is the migration velocity of radionuclide [m/y].

In recent paper, Carlsen *et al.* [16] described a theoretical evaluation of the influence of complexation on radionuclide migration in environments containing an excess of complexing agents. They considered that the equilibrium between the free and complexed metal ions could be regarded to follow pseudo-first order kinetics. They presented an effective retention factor as a controlling factor, which is given as follows:

$$R_f^{\text{eff}} = \frac{R_f(M) + \beta' R_f(ML_i)}{1 + \beta'} \quad (27)$$

where  $\beta'$  is the apparent stability constant of the complex expressed as the ratio between the pseudo-first order rate constant for the formation of the complex and the first order rate constant for the dissociation of the complex, respectively;  $R_f(M)$  is the retention factor for the metal ions in the absence of complexing agents, i. e.,  $\beta' = 0$ ;  $R_f(ML_i)$  is the retention factor for the complex when the apparent stability constant  $\beta'$  approaches

infinity, i. e.,  $\beta' = \infty$ ;  $L_i$  is the average number of ligands per metal ion. However, this effective retention factor is thought to be not rigorous and do not includes detailed complexation mechanisms and related parameters.

In this study, retardation factor equivalent to the retention factor is expressed as follows:

$$R = \frac{R_1}{K_M} + R_2 \quad (28)$$

or

$$R = \left[ \frac{K_M + 1}{K_M} \right] + \frac{(1-\theta) \rho_b}{\theta} \left[ \frac{K_{d1}}{K_M} + K_{d2} \right] \quad (29)$$

If  $K_M \gg 1$ , the first term of the right side of the Eq. (29) will be reduced to 1 and given as more sophisticated form:

$$R = 1 + \frac{(1-\theta) \rho_b}{\theta} K_D \quad (30)$$

where

$$K_D = \frac{K_{d1}}{K_M} + K_{d2} \quad (31)$$

Then, the apparent migration speeds of ionic and chelated radionuclide can be evaluated by the following equation:

$$\frac{V}{R} = \frac{V'}{R'} = \frac{(1+K_M) V_1}{(R_1 + K_M R_2)} \quad (32)$$

Depending upon parameters, the apparent speed of ionic radionuclide in the presence of chelating agents can be greater or less than that of ionic radionuclide without chelating agents. Also the apparent speed of the ionic radionuclide in the presence of chelating agents,  $V'/R$ , is equal to that of the chelated radionuclide,  $V_1/R_1$ .

Since the apparent speed of the ionic radio-

nuclide without chelating agents is given as  $V/R$ , the relationship can be presented as follows:

$$\frac{V}{R} = \frac{V'}{R'} > \frac{V_1}{R_1} \text{ when } \frac{R_2}{R_1} < 1 \quad (33)$$

and

$$\frac{V}{R} = \frac{V'}{R'} < \frac{V_1}{R_1} \text{ when } \frac{R_2}{R_1} > 1 \quad (34)$$

In most cases, since the retardation factor of an ionic radionuclide is much greater than that of chelated radionuclide, Eq. (33) will prevail in actual geologic transport process and this phenomena may be due to the low capacity of the adsorption for the chelated radionuclide. In other words, the apparent speed of a radionuclide in the presence of chelating agents is greater than the case in the absence of the chelating agents. This is well consistent with the reported experimental results [1].

**Numerical calculation**

The calculated results of the solution, Eq.(24), is shown in Fig. 1 and 2 for the numerical illustration of the results of calculation of the model

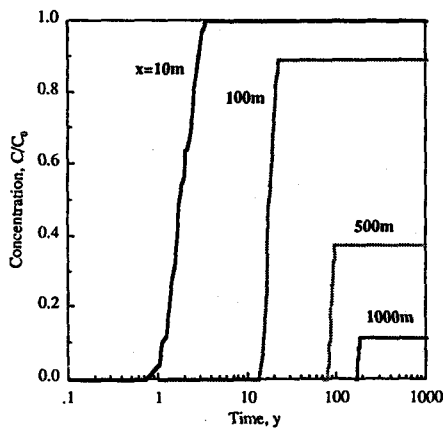


Fig. 1 The calculated results from the solution, Eq(24), with variation of distance.

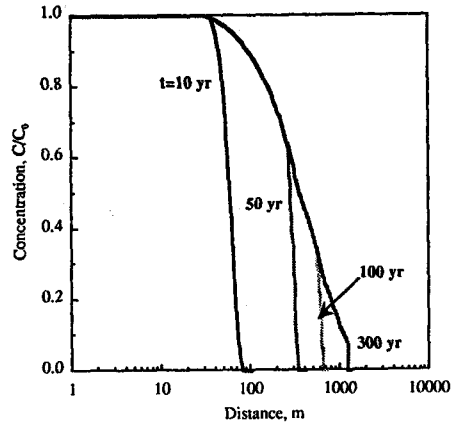


Fig. 2 The calculated results from the solution, Eq.(24), for the various time as a distance from the source increases.

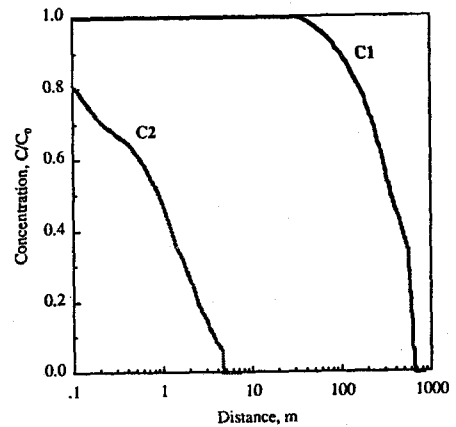


Fig 3. The comparison of the calculated results from the solution, Eq.(24), in the presence of chelating agents(C1) and in the absence of chelating agents (C2), respectively.

with various distances and times, respectively. Table 1 lists the values of parameters used for the calculation of the results. Also comparative results of calculation is showed in Fig. 3 between the case of presence of chelating agent (C1) which incorporating all the mechanisms developed in



**Table 1** The values and notations of parameters used for the numerical calculations

Parameters	Notations	Values
$\theta$	porosity of the porous medium	0.45
$\rho_b(\text{kg/m}^3)$	bulk density of the porous medium	$2.6 \times 10^3$
$\lambda (\text{y}^{-1})$	decay constant of a radionuclide	0.05
$D_1 (\text{m}^2/\text{y})$	dispersion coefficient of a free radionuclide	1.0
$D_2 (\text{m}^2/\text{y})$	dispersion coefficient of a chelated radionuclide	1.0
$V (\text{m}/\text{y})$	velocity of the groundwater in the porous medium	100
$K_M$	apparent chelate formation constant	$10^7$
$k_{dc} (\text{y}^{-1})$	degradation constant of a chelated radionuclide	$10^{-4}$
$K_{d1} (\text{m}^3/\text{kg})$	distribution coefficient of a free radionuclide	$10^5$
$K_{d2} (\text{m}^3/\text{kg})$	distribution coefficient of a chelated radionuclide	$5 \times 10^3$

this study and the case of absence of chelating agents (C2) which is normally considered in previous studies. It is noticed that the presence of chelating agents accelerates the migration rate of radionuclide, i. e., the migration rate of the chelated radionuclide is higher than that of the ionic radionuclide in a porous medium. This may be due to the lower value of the retardation factor of the chelated radionuclide than that of ionic radionuclide.

## CONCLUSIONS

This model study have demonstrated that the effects of chelating agents on the migration of a radionuclide is significant because the chelating agents increase the solubility of the radionuclide for the ground water and decrease the adsorption capacity (i. e., distribution coefficient) of the sorbing geologic medium. The calculated results from the developed model show that the migration rate of the chelated radionuclide is much higher than that of the ionic radionuclide in spite of the

degradation of the chelating agents and the chelated radionuclide.

Since the migration of the chelated radionuclide is much faster than the usual ionic radionuclide, a new remedial action should be developed to reduce the generation and release of chelating agents from the nuclear power plant into the environment. Also a performance assessment for the radioactive waste repository should include the migration of a chelated radionuclide.

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## 다공성 매질에서의 착화하는 방사성핵종의 이동에 대한 이론적 접근

백민훈 · 이건재

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### 요 약

이온 또는 착화된 상태의 방사성핵종의 이동에 대한 착화제의 영향을 조사하기 위하여 새로운 모델이 제시되었다. 착화된 방사성핵종의 이동거동은 착화제와 착화된 방사성핵종의 열화를 포함하는 대류-확산 이동방정식에 의해 해석되었다. 이 수학적 모델은 해석적인 방법에 의해 구해졌으며 지연요소를 조사함으로써 분석되었다. 계산결과들은 감소된 지연요소에 의해 착화된 방사성핵종의 이동속도가 이온형태의 방사성핵종보다 매우 빠름을 보여주었다. 따라서 원자력발전소로부터의 착화제의 발생과 환경으로의 유출을 감소시킬 수 있는 새로운 구제책이 필요하다고 하겠다.