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A Theoretical Study on the Radionuclide Transport Mediated by Pseudo-Colloid in the Fractured Rock Medium

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균열 암반 매질에서 의사콜로이드에 의해 매개된 방사성 핵종의 이동에 대한 이론적 연구

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Abstract

In this study, a transport model was developed in order to analyze and predict the transport behaviors of radionuclides mediated by pseudo-colloid in the fractured rock media. The effect of pseudo-colloid formation on the transport of a radionuclide was shown to be very significant when an apparent pseudo-colloid formation constant, K_{ap}(m^3/kg), was greater than 100. It was resulted from example calculations that the transport of Pu-239 was faster than Ni-63 because pseudo-colloid formation constant of Pu-239 was greater than that of Ni-63. Thus, it can be concluded that acceleration of radionuclide migration may be occurred because the pseudo-colloid formation of radionuclides increases the amount of mobile components in the solution and consequently decreases the amount of radionuclides adsorbed on the stationary solid medium.

요 약

의사콜로이드에 의해 매개된 균열 암반 매질에서의 방사성 핵종의 이동 거동을 분석하고 예측하기 위한 이동모델이 개발되었다. 명목적인 의사콜로이드 형성 상수인 K_{ap}(m^3/kg)값이 100보다 클 때 방사성 핵종의 이동에 매우 큰 영향을 미치는 것으로 나타났다. 계산결과에 의하면 Pu-239의 의사콜로이드 형성상수가 Ni-63보다 크기 때문에 Pu-239의 이동이 Ni-63보다 빠른 것으로 나타났다. 따라서 결론적으로 균열 암반에서의 방사성 핵종의 가속화는 방사성 핵종과 자연성 참콜로이드와의 의사콜로이드 형성에 따른 용액에서의 유동성 성분의 증가와 방사성 핵종들의 정지된 고체매질로의 흡착양 감소에 기인함을 알 수 있다.

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1. Introduction

Until now, most studies on radionuclide transport in groundwater have focused on the transport of dissolved forms of radionuclides. However, it has been reported that a formation of colloidal particles (true radio-colloids) or the sorption of the radionuclides on negatively charged naturally occurring colloidal matters (forming pseudo-colloids) could drastically increase the mobility of the radionuclides released from radioactive wate repository [1−9].

Colloids are omnipresent and abundant in biosphere and geosphere. Most of colloidal particles such as clay minerals, silicic acid, iron hydroxide and organic matter (humic substances) have possibility to transport the radionculides farther and faster than in the absence of them [1−9]. Radionuclides sorbed on these colloidal particles show the same transport characteristics as colloid particles themselves. Radio-colloids are colloids containing radionuclides. They possess all the properties of normal colloids and in addition hold radioactivity. Therefore, it is important to study the roles of colloidal particles in the geologic environment from the view point of the performance and safety assessment of radioactive waste disposal.

In recent years, many models [5, 10−18] have been presented to characterize the behavior of colloids in subsurface systems but there has been no unified approach nor sufficient information to colloid transport modeling. In this study, therefore, we will concentrate on the problem of the pseudo-colloids transport and their role in the transport of radionulide in a fractured rock medium.

2. Theoreical Background

Colloids are characterized as particles containing several hundred to several thousand atoms, ions, or molecules and having diameters ranging from ten to one thousand angstroms [5]. Particles smaller than 1nm(10 angstrom) do not exist as a discrete phase, and any system containing them is not considered heterogeneous [14]. Soil scientists generally regard a diameter of 2000 or 3000nm as an upper limit of particle size for colloids. Besides, the most important properties of colloids are surface interactions such as sorption since colloid has a large surface area (30 0m^2/g) [5].

Olofsson et al. [1] classified radio-colloid as true-colloid and pseudo-colloid according to the processes of colloid formation. True-colloids are formed by condensation of molecules or ions as a result of hydrolytic or precipitation processes [1, 4, 19]. They consist of hydroxides or polymers formed by hydrolysis. They also have a very rapid formation rate and the process is favored at high pH values. Pseudo-colloids, on the other hand, are formed as a result of adsorption on impurities in the solution and tend to be much larger than true-colloids (up to 5000 angstroms) [1]. The formation rate of pseudo-colloids is basically determined by the sorption rate on colloidal impurities. The colloidal impurities could be either inorganic (clays and hydrous oxides) or organic (humic substances).

Many investigations [1−5, 17, 20, 21] have showed that the colloid particles may be very poorly sorbed on geologic media in comparison with ionic or molecular radionuclides. Generally, true-colloids are poorly sorbed on the geologic media such as rock and clay but pseudo-colloids are well sorbed [17].

The roles of colloidal particles in the transport through geosphere have been investigated [5, 10−18]. It was pointed out that colloidal particles could behave as potential vehicles for radionuclides, especially for sorbed actinide elements. Up to date, both laboratory and site studies have demonstrated the existence of colloiadal transport and in some cases the accelerated transport of colloids have been observed [7, 8].

Travis and Nuttall [5] developed a theory for colloid transport using the population balance concept and they also used the multidimensional form of the population balnce equation to directly analyze the effects of polydispersed colloids. The phenomena known as a hydrodynamic chromatography (HDC) has been also used for the description of the colloid transport [15, 16]. In addition, theories and models for predicting colloid migration through porous media have been developed by using filtration theories [10−14].

3. Model Development

3.1. Transport System Formulation

Colloidal particles or dissolved organic matters such as humic substances can act as carriers to enhance the transport of radionulides in groundwater. When either of these materials is present, the transport system can be considered as consisting of three phases :

- aqueous phase (C-phase),
- carrier phase (σ-phase), and
- the stationary solid matrix phase (S-phase).

Radionuclides may be present in either one or all of these phases.

In the followings, we will consider the case of a thin rigid fracture situated in a saturated porous rock (see Fig. 1). For the construction of transport system in a fractured rock medium, we will follow the approach of MIMOSA developed by Lee et al. [22, 23]. The following assumptions are to be introduced :

- colloids are too large to diffuse into the rock matrix,
- advective-dispersive transport can be applied to colloial transport in the fracture,
- the concentration of natural true-colloids is saturated and remains constant, and
- all pseudo-colloids have the same physico-chemical properties.

The assumption that the concentration of the natural true-colloids is saturated and remains constant may be reasonable from the fact that all natural colloids are omnipresent and maintain at constant concentration in every depths of natural groundwater systems [21].

Therefore, the formulation of transport equations in the presence of colloids will be done considering the following processes :

- advective transport of pseudo-colloids and radionuclides along the fractrue,
- longitudinal mechanical dispersion of pseudo-colloids and radionuclides in the fracture,
- molecular diffusion of pseudo-colloids and radionuclides within the fracture along the fracture axis,
- molecular diffusion of radionuclides from the fracture into the rock matrix,
- sorption of colloids and radionuclides onto the surface of the fractrue,
- sorption of radionuclides within the rock matrix,
- sorption of radionuclides onto the natural true-colloidal surface (pseudo-colloid formation),
- radioactive decay of radidonuclides, and
- filtration of pseudo-colloids due to interactions with fracture surface.

In our transport system, as shown in Fig. 1, there are six components consisting of :

(1) dissolved mobile radionuclide R(C_r, kg/m^3)

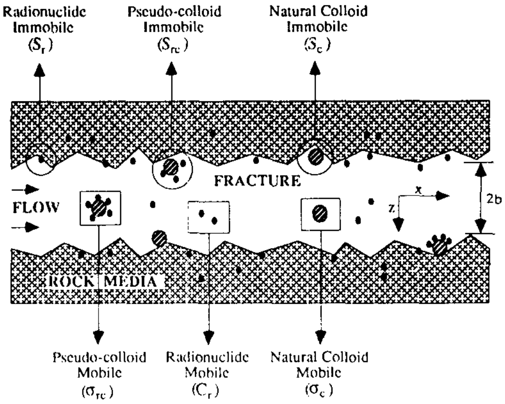

Fig. 1. Colloidal Transport System in the Fracture and Porous Rock Matrix.

(2) saturated mobile natural true-colloid C(σ^*_c, kg/m^3)
(3) mobile radio-or pseudo-colloid RC((σ^*_{rc}, kg/m^3)
(4) immobile radionuclide SR(S_r, kg/m^2)
(5) saturated immobile natural true-colloid SC(S^*_c, kg/m^2)
(6) immobile pseduo-colloid SRC (S^*_{rc}, kg/m^2)

First of all, consider a radionuclide R at concentration $C_r(x, t)$ in groundwater in the fracture. Also present in the fracture are natural true-colloids maintained at saturated equilibrium concentration σ^*_c(constant). The radionuclide R will sorb onto the natural true-colloid C to form a radio- or pseudo-colloid RC with concentration of $\sigma_{rc}(x, t)$. The three components present in the fracture groundwater will be also sorbed onto the stationary solid phase and form three immobile components, SR, SC, and SRC, respectively. However the immobile component SC will not vary with time and distance because the equilibrium concentration of C in the solution was assumed to be saturated and remain constant. Therefore, the concentrations of the immobile components can be expressed as $S_r(x, t)$, S^*_c, and $S_{rc}(x, t)$, respectively.

3.2. Basic Equations

Pseudo-colloid Formation

For the pseudo-colloid formation process, we assume that adsorption of a radionuclide R on the natural colloidal surfaces is instantaneous and adsorption sites on the mobile and immobile colloids are in equilibrium with aqueous phase. However the amount of radionculides adsorbed on the colloids will depend upon the amount of colloids which are available in the groundwater. Thus we can consider the sorption process as a chemical reaction similar to a complex formation. Accordingly the sorption process can be expressed as :

$$R + C \leftrightarrow RC \qquad (1)$$

The equilibrium relationship between R and C (i.e., relationship between C_r and σ_{rc}) can be described as the following equilibrium equation :

$$K_p = \frac{\sigma_{rc}}{C_r \sigma_c}. \qquad (2)$$

where K_p(m^3/kg) is the equilbrium formation constant of the pseudo-colloid RC.

However, since the concentration of the natural true-colloid is constant, the above relationship must be revised. Thus a new equilibrium constant named as an apparent pseudo-colloid formation constant is introduced to consider the constant saturated concentration of the natural true-colloids :

$$K_{ap} = \frac{\sigma_{rc}}{C_r \sigma^*_c} \qquad (3)$$

or

$$\sigma_{rc} = K_{ap} C_r \sigma^*_c . \qquad (4)$$

where K_{ap}(m^3/kg) is the apparent pseudo-colloid formation constant.

Filtration Process

A general equation for the mechanism of filtration has been proposed by Tien et al. [10] and Harvey et al. [13] among others. In the fracture, filtration can occur through the pores within fracture filling materials and on the surfaces of the fracture because colloids are too large to diffuse into the porous rock matrix. Filtration rate for the mobile pseudo-colloid RC and be given as [11, 14] :

$$F_{rc}(x, t) = \frac{\partial S_{rc}}{\partial t} = \lambda_{frc} V \sigma_{rc} \qquad (5)$$

where λ_{frc} is the filtration coefficient of the pseudo-colloid RC (1/m) and V is the average groundwater velocity in the fracture (m/yr)

Hydrodynamic Dispersion Coefficient

In the case of uniform flow and an isotropic medium, the hydrodynamic dispersion coefficient for the dissolved radionuclide R can be expressed as [24] :

$$D_r = \alpha_r V + D_r^* \qquad (6)$$

where α_r is the dispersivity of the radionuclide R in the direction of the fracture axis (m) and D^*_r is the molecular diffusion coefficinet of the radionuclide R in groundwater (m^2/yr).

However, in the case of a colloid or pseudo-colloid, the hydrodyanmic dispersion coefficient differs from that of the radionuclide since the Brownian diffusion coefficient for RC, D_{Brc}, is not necessarily equal to the molecular diffusion coefficient of the radionuclide R, D^*_r. Thus hydrodynamic dispersion coefficient of a pseudocolloid RC can be expressed as [18]:

$$D_{rc} = \alpha_{rc} V + D_{Brc} \tag{7}$$

where α_{rc} is the disperisvity of the radio-colloid RC in the direction of the fracture axis (m) and D_{Brc} is the Brownian diffusion coefficient of the pseudo-colloid RC in the groundwater (m^2/yr).

Sorption Process

The use of a simple transport model, with linear sorption both on pore surfaces or fracture surfaces and on colloids, is desirable if it can be demonstrated to be conservative, firstly because the lack of comprehensive data prohibits extensive use of more complex isotherms and secondly because the resulting system of linear transport equation is straightforward to solve in comparison with those arising from non-linear sorption [20]. Several investigators suggested that a simplified linear sorption model for colloid transport can be used conservatively [20, 21].

Thus it is assumed that sorption of radionuclide R and pseudo-colloid RC on fracture surfaces is governed by a reversible equilibrium linear sorption isotherm using the concept of distribution coefficient.

$$S_r = K_{dr} C_r \tag{8}$$

$$S_{rc} = K_{drc}\, \sigma_{rc} \tag{9}$$

where K_{dr} (,) and K_{drc} (m) are the distribution coefficients of the radionuclide R and pseudo-colloid RC, respectively.

However these relationships should be corrected in order to consider multicomponent effects in this sorption system because the distribution coefficients K_{dr} and K_{drc} would be changed when the two components R and RC are present simultaneousey and affecting each other. Thus new relationships, in the presence of colloids, can be presented as:

$$K^*_{dr} = \frac{S_r}{C_r + \sigma_{rc}} = \frac{K_{dr}C_r}{\left(1 + K_{ap}\sigma^*_c\right)C_r} = \frac{1}{1 + K_{ap}\sigma^*_c}\, K_{dr} \tag{10}$$

and

$$K^*_{drc} = \frac{S_{rc}}{C_r + \sigma_{rc}} = \frac{K_{drc}\sigma_{rc}}{\left(1 + \frac{1}{K_{ap}\sigma^*_c}\right)\sigma_{rc}} = \frac{K_{ap}\sigma^*_c}{1 + K_{ap}\sigma^*_c}\, K_{drc} \tag{11}$$

where K^*_{dr} (m) and K^*_{drc} (m) can be called as effective distribution coefficients of the radionuclide R and pseudo-colloid RC, respectively.

Diffusive Loss

In the case of fractured media, colloids may be excluded from matrix pores because colloids are too large to diffuse into the rock matrix, in which case radionuclides bond to them are not subjected to the retarding effects of matrix diffusion and sorption onto matrix pore surfaces [20]. Therefore, for the diffusive loss flux, radionuclide R only diffuses into the rock matix porous rock matrix with bulk density ρ_b (kg/m^3) crossing the fracture-matrix interface. Generally this diffusive flux, q_r ($kg/m^2/yr$) can be expressed by Fick's first law:

$$q_r(x, t) = -\phi_d D_{pr} \left.\frac{\partial C_{pr}(x, z, t)}{\partial z}\right|_{z=b} \qquad x > 0,\ t > 0 \tag{12}$$

where ϕ_d is a diffusion porosity of the rock matrix, D_{pr} is the effective diffusion coefficient of the radionuclide R in the rock matrix (m^2/yr), and $C_{pr}(x, z, t)$ is the concentration of the radionuclide R in the rock matrix (kg/m^3).

3.3. Governing Equations

Governing equations for the radionuclide R and pseudo-colloid RC in the fracture and rock matrix will be established from the basic equations and appropriate mass balances. The processes in the transport system of the Fig. 1 can be described by three coupled, one dimensional partial differential equations, two for the radionuclide and pseduo-colloid in the fracture and one for the radionculide in porous rock matrix. The coupling between the fracture and porous rock media is provided by the continuity of fluxes and concentrations along the interface.

Transport Equation in the Fracture

The differential advective-dispersive transport equation for the radionuclide R in the fracture with half aperture b (m) can be given as :

$$\frac{\partial C_r}{\partial t}+\frac{1}{b}\frac{\partial S_r}{\partial t}=D_r\frac{\partial^2 C_r}{\partial x^2}-V\frac{\partial C_r}{\partial x}$$

$$-\lambda_{dr}\left(C_r+\frac{1}{b}S_r\right)-\frac{q_r}{b} \qquad (13)$$

$$x>0,\quad t>0$$

The differential advective-dispersive transport equation for the pseudo-colloid RC in the fracture can be also given as :

$$\frac{\partial \sigma_{rc}}{\partial t}+\frac{1}{b}\frac{\partial S_{rc}}{\partial t}=D_{rc}\frac{\partial^2 \sigma_{rc}}{\partial x^2}-V\frac{\partial \sigma_{rc}}{\partial x}$$

$$-\lambda_{dr}\left(\sigma_{rc}+\frac{1}{b}S_{rc}\right)-\lambda_{frc}V\sigma_{rc} \qquad (14)$$

$$x>0,\quad t>0$$

Combining Eqs. (13) and (14), using the Eqs. (4), (10), (11), and (12) the governing equation for the total amount of radionuclide in the fracture can be given as :

$$R^*\frac{\partial C_r}{\partial t}=D^*\frac{\partial^2 C_r}{\partial x^2}-V^*\frac{\partial C_r}{\partial x}-\lambda_{dr}R_T C_r$$

$$-\frac{\phi_d D_{pr}}{b}\left.\frac{\partial C_{pr}}{\partial z}\right|_{z=b} \qquad (15)$$

where

$$R^*=1+\frac{K_{dr}^*}{b}+\frac{K_{drc}^*}{b} \qquad (16)$$

$$D^*=D_r+K_{ap}\sigma_c^*D_{rc} \qquad (17)$$

$$V^*=\left(1+K_{ap}\sigma_c^*\right)V \qquad (18)$$

$$R_T=R^*+\frac{\lambda_{frc}\,V\,K_{ap}\,\sigma_c^*}{\lambda_{dr}}. \qquad (19)$$

Transport Equation in the Rock Matrix

The differential mass balance equation for the radionuclide R in the porous rock matrix can be obtained in a similar way by considering the mass balance for a strip of unit width, extending in the direction perpendicular to the fracture axis.

$$\phi_d\frac{\partial C_{pr}}{\partial t}+\rho_b\frac{\partial S_{pr}}{\partial t}=\phi_d D_{pr}\frac{\partial^2 C_{pr}}{\partial z^2}$$

$$-\lambda_{dr}\left(\phi_d C_{pr}+\rho_b S_{pr}\right) \qquad b\le z\le\infty \qquad (20)$$

where S_{pr} (x, z, t) is the mass of radionuclides adsorbed per unit mass of solid in the porous rock matrix (kg/kg) and ρ_b is the bulk density of the rock matrix (kg/m^3). The effective diffusion coefficient D_{pr} is defined here as :

$$D_{pr}=\tau D_r^* \qquad (21)$$

where τ is a matrix tortuosity.

For adsorption within the rock matrix a linear equilibrium isotherm is assumed as :

$$S_{pr}=K_{dpr}C_{pr} \qquad (22)$$

where K_{dpr} (m^3/kg) is the distribution coefficient of the radionuclide R in the porous rock matrix defined as the mass of the R adsorbed per unit volume of solid divided by the concentration of R in the solution.

Therefore, the final governing equation for the radionuclide R in the rock matrix is given by substituting the differtial form of the Eq. (22) into the Eq. (20):

$$R_{pr}\frac{\partial C_{pr}}{\partial t} = D_{pr}\frac{\partial^2 C_{pr}}{\partial z^2} - \lambda_{dr}R_{pr}C_{pr} \quad (23)$$

where

$$R_{pr} = 1 + \frac{\rho_r}{\phi_d}K_{dpr} \quad (24)$$

Solutions of the Governing Equations

Initial and boundary conditions for the governing equations (15) and (23) are given as follows:

$$C_r(x, 0) = 0 \quad (25a)$$

$$C_r(0, t) = C_0 \exp(-\lambda_{dr}t) \quad (25b)$$

$$C_r(\infty, t) = 0 \quad (25c)$$

$$C_{pr}(x, z, 0) = 0 \quad (25d)$$

$$C_{pr}(x, b, t) = C_r(x, t) \quad (25e)$$

$$C_{pr}(x, \infty, t) = 0 \quad (25f)$$

The solutions of the governing equations (15) and (23), subject to the initial and boundary conditions Eq. (25), can be obtained by applying the Laplace transform to the Eqs. (15) and (23) and applying the concentration gradient of the radionuclide at the fracture-rock matrix interface, $\frac{\partial C_{pr}}{\partial_z}\mid_{z=b}$, which can be obtained from the Eq. (23), to the Eq. (15) [22]/ Therefore, the analytic solution of the Eq. (15) for the radionuclide in the fracture subject to the conditions Eq. (25) is given simply as:

$$\frac{C_r}{C_0} =$$

$$\frac{2}{\sqrt{\pi}}\exp(-\lambda_{dr}t)\exp(Vx)\int_l^{\infty}\exp\left[-\xi^2 - \frac{x^2}{4\xi^2}(v^2+\kappa)\right]$$

$$\times\ \mathrm{erfc}\left[\frac{1}{2\sqrt{t - {\beta x^2}/{4\xi^2}}}\left(\frac{A x^2}{4\xi^2}\right)\right]d\xi \quad (26)$$

where

$$l = \frac{x}{2}\left(\frac{R^*}{D^* t}\right)^2 \quad (27)$$

$$v = \frac{V^*}{2D^*} \quad (28)$$

$$\eta = \sqrt{\frac{R_{pr}}{D_{pr}}} \quad (29)$$

$$A = \frac{\phi_d D_{pr}}{b D^*}\eta \quad (30)$$

$$\beta = \frac{R^*}{D^*} \quad (31)$$

$$\kappa = \frac{\lambda_{dr}(R_T - R^*)}{D^*} \quad (32)$$

For the radionuclide in the rock matrix, the analytic solution is given as:

$$\frac{C_{pr}}{C_0} =$$

$$\frac{2}{\sqrt{\pi}}\exp(-\lambda_{dr}t)\exp(Vx)\int_l^{\infty}\exp\left[-\xi^2 - \frac{x^2}{4\xi^2}(v^2+\kappa)\right]$$

$$\times\ \mathrm{erfc}\left[\frac{1}{2\sqrt{t - {\beta x^2}/{4\xi^2}}}\left(\frac{A x^2}{4\xi^2} + \eta(z-b)\right)\right]d\xi \quad (33)$$

On the other hand, the fractional concentration of the pseudo-colloid RC is given by using the relationship Eq. (4), i.e.,

$$\frac{\sigma_{rc}}{C_0} = \frac{2K_{ap}\sigma_c^*}{\sqrt{\pi}}\exp(-\lambda_{dr}t)$$

$$\exp(Vx)\int_l^{\infty}\exp\left[-\xi^2 - \frac{x^2}{4\xi^2}(v^2+\kappa)\right]$$

$$\times\ \mathrm{erfc}\left[\frac{1}{2\sqrt{t - {\beta x^2}/{4\xi^2}}}\left(\frac{A x^2}{4\xi^2}\right)\right]d\xi \quad (34)$$

4. Example Calculations and Discussion

Example calculations were performed in order to analyze and simulate the transport phenomena of raidonulides in the presence of colloids. For the example calculations, geohydrological parameters involved in the developed transport system are listed in Table 1. Standard values of paramters related with radionuclides (i.e., Tc-99, Pu-239, and Ni-63) and their pseudo-colloids are also listed in Table 2 in order to analyze the transport behaviors of the radionucolides in the fractured media. The values of K_{ap} and σ_c^* were obtained from literature [21], however, other values are assumed for a common crystalline rock medium such as granite.

Colloids in crystalline rock systems such as granite are basically composed of components of aquifer materials (clay, silica). Under the hydrogeochemical conditions existing at depth in crystalline rock systems, colloid concentrations are not expected to exceed 100 ppb [6]. However under transient conditions when physical and/or chemical changes are occurring (e.g., hydrothermal activity), colloid generation may be enhanced [6]. Recently Allard et al. [21] presented the assessed concentrations of particulate matters such as inorganic colloids, micro-organisms, and humic substances in Swedish deep groundwaters (see Table 3). We selected the value of σ_c^* as somewhat greater than presented in Table 3 in order to assess conservatively.

Table 1. Hydrogeochemical Parameters Used in Example Calculations

Parameters	Value	Unit
b	0.0001	m
V	10	m/yr
ϕ_d	0.1	–
ρ_r	2700	kg/m³
α	0.5	m
τ	1.0	–
D_r^*	0.001	m²/yr
D_{Brc}	0.001	m²/yr

Allard et al. [21] also presented sorption coefficient for the particulate matter K_{part} (m^3/kg), similar to the pseudo-collioid formation constant K_p presented in this study:

$$K_{part} = \frac{C_s}{C \cdot S} \tag{35}$$

where C_s (kg/m^3) is the concentration of a nuclide attached to the particulate matter C (kg/m^3) is the concentration of a nuclide in the water, and S (kg/m^3) is the concentration of particulate matter in the water. In this study we referred the values of K_{part} presented by Allard et al. [21] in the selection of K_{ap} listed in Table 2.

Also we assumed the Brownian diffusion coefficient of pseudo-colloid, D_{Brc}, to be equal with the molecular diffusion coefficient of nuclide, D_r. How-

Table 3. Assessed Concentrations of Colloids and Microorganisms as Well as Humic Substances in Groundwaters [21]

Concentrations, $(kg/m^3) \cdot 10^6$	Central Value	Max. Value
Inorganic colloids	100	400
Humic Substances (HA, FA)	100	500
Microorganisms	10	50

HA = humic acid ; fulvic acid

Table 2. Standard Values of Paramters Related with Radionuclides

Radio-nuclide	λ_{dr} (1/yr)	K_{dr} (m)	K_{dpr} (m^3/kg)	K_{ap} (m^3/kg)	K_{drc} (m)	λ_{frc} (1/yr)	σ_c^* (kg/m^3)
Tc-99	3.3007E-6	0	0	30	0	0	5E-3
Pu-239	2.8881E-5	4E-5*	0.5	30	0	0	5E-3
Ni-63	6.9315E-3	1.3E-5	0.15	1	0	0	5E-3

* $4E\text{-}5 = 4.0 \times 10^{-5}$

ever, as a first approximation, it has been presented that the ratio of colloid dispersivity to solute disperpersivity in simple fractures can be given by

$$\frac{1}{\eta}=\frac{\text{colloid radius}}{\text{ionic radius}} \tag{25}$$

Calculation results using the Tables 1 and 2 are shown in Fig. 2 for each of radionuclides with varying time. With varying distance, calculation results for each of radionuclides are given in Fig. 3 by using the data in Tables 1 and 2. Among the three radionuclies, as shown in Figs. 2 and 3, the transport of Tc-99 is fastest in the fractured rock matrix. This may be due to the fact that Tc-99 is nearly not adsorbed on the solid medium. In the case of Pu-239 and Ni-63, which have similar capacities of adsorption, the transport of Pu-239 is faster than Ni-63 because pseudo-colloid formation constant of Pu-239 is greater than that of Ni-63.

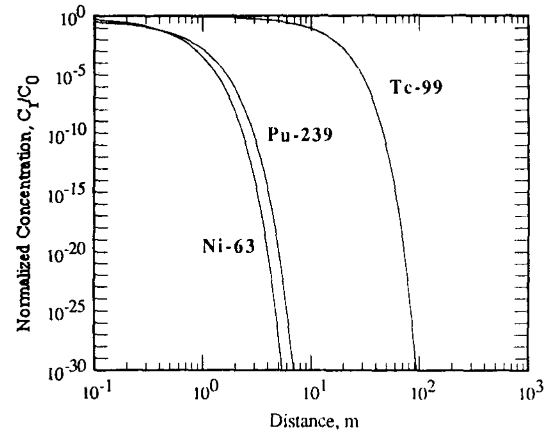

Fig. 3. Normalized Concentrations of te Three Radionuclides at t=100 yrs with Varying Distance.

The effect of apparent pseudo-colloid formation constant, K_{ap}, on the transport of a radionuclide is analyzed in Fig. 4 using the data listed in Tables 1 and 4. It is shown that the released concentration or transport rate increases as the values of K_{ap} increase. It is also noticed that the effect of pseudo-colloid formation on the transport in not negligible when

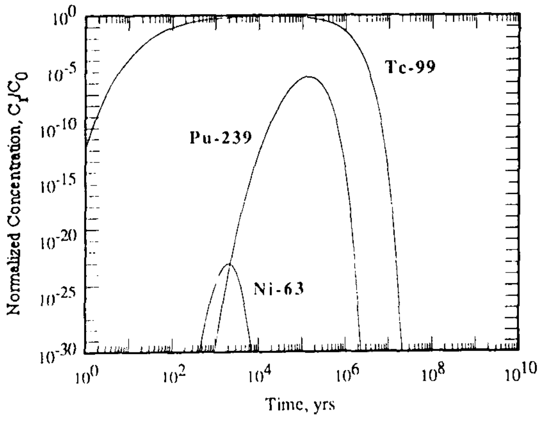

Fig. 2. Normalized Concentrations of the Three Radionculides at x=10m with Varying Time.

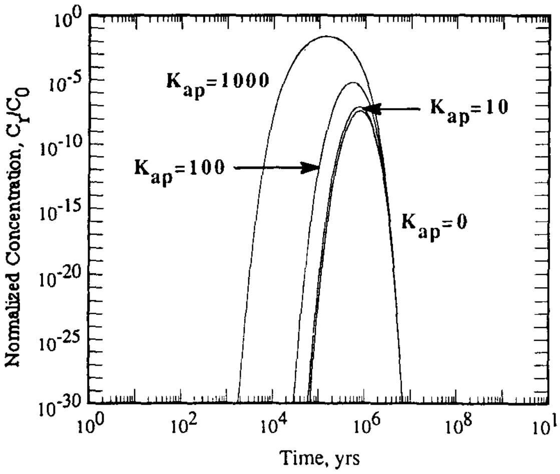

Fig. 4. Effect of Pseudo-Colloid Formaion Constant K_{ap} at x=300m with Varying Time.

Table 4. Parameters Used to Analyze the Effect of K_{ap}

x (m)	λ_{dr} (1/yr)	K_{dr} (m)	K_{dpr} (m^3/kg)	K_{drc} (m)	λ_{frc} (1/yr)	σ_c^* (kg/m^3)
300	1×10^{-5}	1×10^{-5}	1×10^{-3}	1×10^{-5}	1×10^{-3}	1×10^{-3}

$10 < K_{ap} < 100$. When $K_{ap} > 100$, the effect of pseudo-colloid formation on the transport of a radionuclide is shown to be very significant.

Recently Grindrod et al. [25] developed a numerical code (COLLAGE) for radionuclide migration through a fractured geosphere in aqueous and colloidal phases. In this code, adsorption behavior is represented by two partition coefficients K_1(dimensionless) and K_2 (dimensionless). K_1 represents the ratio of the density of radionuclides sorbed to free colloidal material, to the density of the dissolved species. Similarly, K_2 represents the ratio of the density of radionuclides sorbed to immobile colloidal material to the density of the dissolved species. In this code, however, filtration was not considered. The example calculation output from the COLLAGE is shown in Fig. 5 with varying K_1's(dimensionless). Fig. 5 shows that nuclide concentration released is increased about three orders of magnitude when K_1 increases from 0 to 50. This is somewhat greater value than that of predicted in our study (see Fig. 4). However it can be noted that the results presented in Figs. 4 and 5 show similar behaviors despite of different transport models and parameters used.

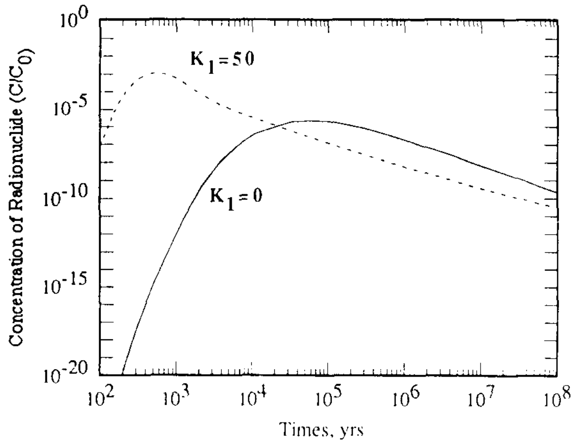

Fig. 5. Example Output From the COLLAGE Program with Varying Colloid formation parameter K_1 (dimensionless).

Other parameters such as distribution coefficient of a radionuclide, K_{dr}, will influence the transport behaviors of radionuclides in the fractured rock media. The effect of colloids on the K_{dr} can be easily analyzed by the Eq. (10). In other words, the distribution coefficent of a radionuclide in the fracture, K_{dr}, will be reduced by a factor of $1/1+K_{ap}\sigma_c^*$ due to the presence of colloids with concentration of σ_c^* and formation constant of K_{ap}.

Similarly Allard et al. [21] estimated the effect of colloids on the sorption of a nuclide using the effective sorption capacty, K_{eff} (m^3/kg), of the nuclide on the rock given as :

$$K_{eff} = \frac{K_d}{1+\sum S \cdot K_{part}} \tag{36}$$

where K_d(m^3/kg) is sorption coefficient on rock and the summation is made over all the particulate species. Thus it is noticed that the Eq. (36) is exactly consistent with the Eq. (10) presented in this study.

5. Conclusions

Following conclusions can be made from this model study :

- Transport behaviors of radionuclides mediated by pseudo-colloid in the fractured rock media could be analyzed and predicted by the transport model developed in this study.
- Acceleration of radionuclides in a fractured rock medium occurs because pseudo-colloid formation of radionuclides with natural true-colloids increases the amount of mobile components in the solution and decreases the amount of radionuclides adsorbed on the stationary solid medium.
- Exisitng information is sufficient to raise concerns about the potential effect of colloids on the accelerated transport of radionuclides. Nevertheless, the effect of colloids on tranport of radionuclides has been underestimated or neglected because of lack of information on the colloid-facilitated transport in sefety assessment models that predict radio-

nuclide transport in the subsurface.

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