Proceedings of the Korean Nuclear Society Spring Meeting

Ulsan, Korea, May 1995

Radionuclide Transport Mediated by Pseudo-Colloid in the Fractured Rock Media : Model Development

Min Hoon Baik and Phil Soo Hahn

Korea Atomic Energy Research Institute

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. It was resulted 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. Also, the effect of pseudo-colloid formation on the transport of a radionuclide was shown to be very significant when the apparent pseudo-colloid formation constant, $K_{ap}(m^3/kg)$, was greater than 100. 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.

1. Introduction

Colloids are omnipresent and abundant in biosphere and geosphere. Most of colloidal particles such as clay minerals, silicic acid, iron hydroxide and humic substances have possibility to transport the radionuclides farther and faster than in the absence of them [1-5].

In recent years, many models [6-10] 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 effect on the transport of radionuclides in the fractured rock media.

2. Model Development

Following assumptions are to be introduced:

- colloids are too large to diffuse into the rock matrix, and
- the concentration of natural colloids is saturated and remains constant.

Since the concentration of the natural colloid C is assumed to be constant ($\sigma_c^* =$ constant), an equilibrium constant named as apparent pseudo-colloid formation constant can be defined as [11]:

$$K_{ap} = \frac{\sigma_{rc}}{C_r \sigma_c^*}, \tag{1}$$

where C_r is the concentration of a dissolved radionuclide R (kg/m^3), σ_c^* is the concentration of the natural colloid C (kg/m^3), σ_{rc} is the concentration of a mobile pseudo-colloid RC (kg/m^3), and K_{ap}(m^3/kg) is the apparent pseudo-colloid formation constant.

Also equilibrium sorption relationships can be given by considering multicomponent effect in our sorption system [11].

$$K_{dr}^* = \frac{S_r}{C_r + \sigma_{rc}} = \frac{K_{dr} C_r}{\left(1 + K_{ap}\sigma_c^*\right) C_r} = \frac{1}{1 + K_{ap}\sigma_c^*} K_{dr} \tag{2}$$

and

$$K_{drc}^* = \frac{S_{rc}}{C_r + \sigma_{rc}} = \frac{K_{drc}\sigma_{rc}}{\left(1 + \frac{1}{K_{ap}\sigma_c^*}\right)\sigma_{rc}} = \frac{K_{ap}\sigma_c^*}{1 + K_{ap}\sigma_c^*} K_{drc} \tag{3}$$

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

A final goveming equation for the total amount of radionuclides in the fracture can be given by using the relationships of Eqs. (1), (2), and (3), and rearranging the resulting equation for C_r.

$$R^* \frac{\partial C_r}{\partial t} = D^* \frac{\partial^2 C_r}{\partial x^2} - V^* \frac{\partial C_r}{\partial x} - \lambda_{dr} R_T C_r - \frac{\phi_d D_{pr}}{b} \frac{\partial C_{pr}}{\partial z}\bigg|_{z=b}, \tag{4}$$

$$R^* = 1 + \frac{K_{dr}^*}{b} + \frac{K_{drc}^*}{b} \tag{5}$$

$$D^* = D_r + K_{ap} \sigma_c^* D_{rc} \tag{6}$$

$$V^* = \left(1 + K_{ap} \sigma_c^*\right) V \tag{7}$$

$$R_T = R^* + \frac{\lambda_{frc} V K_{ap} \sigma_c^*}{\lambda_{dr}} \tag{8}$$

where D_r and D_{rc} are hydrodynamic dispersion coefficients of the radionuclide and the pseudo-colloid (m^2/yr), respectively, V is an average groundwater velocity (m/yr), λ_{dr} is the decay constant of the radionuclide R (1/yr), λ_{frc} is the filtration coefficient of the pseudo-colloid RC through the fracture (1/m), b is the fracture half width (m), and q_r is the diffusive loss flux of the radionuclide R into the porous rock matrix ($kg/m^2/yr$).

Therefore, analytic solution of the Eq. (9) for the radionuclide in the fracture subject to appropriate initial and boundary conditions can be given by considering the diffusive equation for the radionuclide R in the rock matrix with linear sorption isotherm [12].

$$\frac{C_r}{C_0} = \frac{2}{\sqrt{\pi}} \exp(-\lambda_{dr} t) \exp(\nu x) \int_l^\infty \exp\left[-\xi^2 - \frac{x^2}{4\xi^2}(\nu^2 + \kappa)\right]$$

$$\times \operatorname{erfc}\left[\frac{1}{2\sqrt{t - \beta x^2/4\xi^2}} \left(\frac{A x^2}{4\xi^2}\right)\right] d\xi \tag{11}$$

where

$$l = \frac{x}{2}\left(\frac{R^*}{D^* t}\right)^2 \qquad \nu = \frac{V^*}{2 D^*} \qquad \eta = \sqrt{\frac{R_{pr}}{D_{pr}}}$$

$$A = \frac{\phi_d D_{pr}}{b D^*} \eta \qquad \beta = \frac{R^*}{D^*} \qquad \kappa = \frac{\lambda_{dr}(R_T - R^*)}{D^*} \tag{12}$$

3. Results and Discussion

Geohydrological parameters involved in the developed transport system are listed in Table 1 for example calculations. Standard values of the parameters related with radionuclides (i.e., Tc-99, Pu-239, and Ni-63) and their pseudo-colloids are also listed in Table 2. Calculation results using the Tables 1 and 2 are shown in Fig. 1 for each of the radionuclides with varying time. Among the three radionuclides, as shown in Fig. 1, 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

The effect of the apparent pseudo-colloid formation constant, K_{ap}, on the transport of a radionuclide is analyzed in Fig. 2. It is shown that the releasing concentration and transport rate increases as the values of K_{ap} increase. It is also noticed that the effect of pseudo-colloid formation on the transport is not negligible when $10 < K_{ap} < 100$. When $K_{ap} > 100$, the effect of pseudo-colloid formation on the transport of a radionuclide is shown to be very significant.

4. Conclusions

Acceleration of radionuclides in a fractured rock medium occurs because pseudo-colloid formation of radionuclides with natural colloids increases the amount of mobile components in the solution and decreases the amount of radionuclides adsorbed on the stationary solid medium. The effect of colloids on transport of radionuclides has been underestimated or neglected because of lack of information and difficulties of mathematical treatment on the colloid-facilitated transport mechanisms. Thus we should develop a capability to include the colloid-facilitated transport in safety assessment models that predict radionuclide transport in the subsurface.

References

1. U. Olofsson et al., *Scientific Basis for Nuclear Waste Management*, **4**, 191 (1982).
2. A. Avogadro and G. De Marsily, *ibid*, **6**, 495 (1983).
3. J. I. Kim et al., *ibid*, **7**, 31 (1984).
4. C. Degueldre, *ibid*, **16**, 817 (1993).
5. K. A. Orlandini et al., *Environ. Sci. Technol.*, **24**(5), 706 (1990).
6. A. Saltelli et al., *Nuclear Technology*, **67**, 245 (1984).
7. L. M. McDowell-Boyer et al., *Water Resour. Research*, **22**(13), 1901 (1986).
8. R. W. Harvey and S. P. Garabedian, *Environ. Sic. Technol.*, **25**, 178 (1991).
9. S. Nagasaki e t al., *J. of Nuclear Science and Technology*, **31**(2), 143 (1994).
10. G. G. Eichholz et al., *Nuclear Technology*, **58**, 511 (1982).
11. 한필수 외, 처분안전성 연구, 제 2권 처분안전성 평가 코드 개발, KAERI-NEMAC/RR-117/94, 한국원자력연구소, 1994.
12. Y. M. Lee et al., *J. of the Korean Nuclear Society*, **22**(3), 304 (1990).

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/m^3
α	0.5	m
τ	1.0	-
D_r^*	0.001	m^2/yr
D_{Brc}	0.001	m^2/yr

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-5 = 4.0 x 10^{-5}

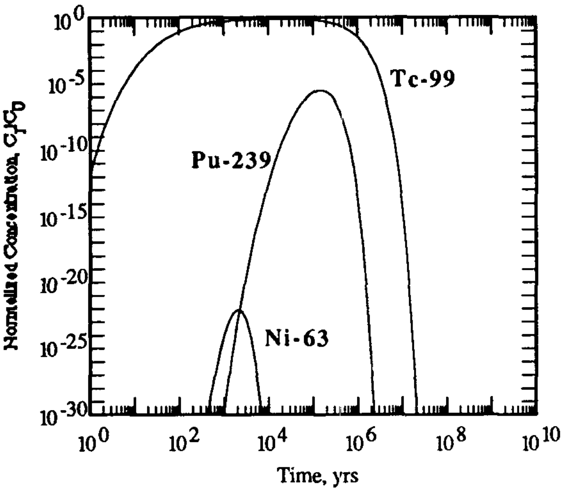

Fig. 1. Normalized concentrations of the three radionculides at x = 10 m with varying time.

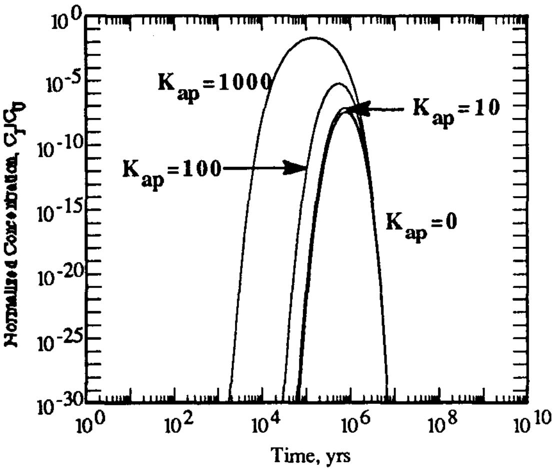

Fig. 2. Effect of pseudo-colloid formaion constant K_{ap} at x=300m with varying time.