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The Pseudo Colloid Migration by the Four-Member Decay Chain in the Fractured Porous Medium

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1. Introduction

In an assessment for the long-term performance of a deep geologic repository in crystalline rocks such as the granite, fractures have been considered as the main pathways of groundwater flow and radionuclide transport. Many papers have already dealt with the problem of the radionuclide transport in various fractured porous matrix, but without discussing daughter products. However, the daughter products may travel much farther than their parents along the fracture. Therefore, some models neglecting the effect of the decay chain may underestimate the transport radionuclide concentration in geological media. We need to understand radionuclide transport process fractured rock and to validate numerical codes used to assess the performance of the potential geologic repository for high-level nuclear waste. The potential repository site, such as Yucca Mountain in USA, much of which is comprised of a fractured volcanic tuff, has led to an increased interest in the behavior of radionuclide transport in fracture.

It is important not only the transport of radionuclides but also the migration of the colloid in the groundwater. Although the size of a colloid is from a hundreds nanometer to a centimeter, that is smaller than the pore. The colloid is not diffused into the surrounding rock matrix, however, solutes can be diffused into the surrounding rock matrix. Although the colloid is heavy, the velocity of the colloid is faster than the solute. As the colloid moves along a fracture, the colloid reaches quicker than the solute to the biosphere.

According to the electromagnetic interaction between the solute and the colloid, solutes are absorbed by the colloid, and then we are called the pseudo-colloid.

The natural colloid can exist inside a fracture with a density of 10^5 particles per one liter of a liquid. When the radionuclide migrates through a fractured rock, solutes sorb on natural colloids as well as the stationary fracture wall solid surface. Due to natural colloids, whose particle size is larger than that of solutes, colloids can migrate faster than solutes. Therefore, these pseudo-colloids, which are the sorbed solute molecules on the natural colloids, can also migrate faster than the solute. Both the solute and the pseudo-colloid are sorbed onto and desorbed from stationary solid wall surface of a fracture. Even though the colloid can not penetrate into the surrounding rock matrix, the solute diffuses into the porous matrix.

This study extends the previous studies to consider the four-member radioactive decay chain and arbitrary inlet boundary conditions with the time-dependent concentration.

2. Mathematical Concepts

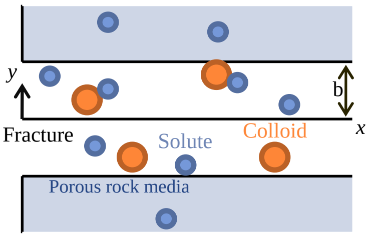

Fig.1. The natural colloid and solutes migration in a fractured porous media.

We consider a radioactive solute at concentration $C_{2i}(x,t)$ in water in a thin fracture of the fractured porous rock. Also present in the fracture are the natural colloids, on which the solute can sorb to form the pseudo-colloids as shown in Fig. 1. Assume the one-dimensional convective-diffusive transport within the fracture, and assume that the colloids are too large to diffuse into the rock matrix. Neglecting the possible colloid filtration within a fracture, the governing equation of the pseudo-colloid migration is:

$$\varepsilon_1\xi_1\frac{\partial C_{1i}(x,t)}{\partial t}+\varepsilon_1\xi_1 v_{1i}\frac{\partial C_{1i}(x,t)}{\partial x}+\varepsilon_1 S_1(x,t)+\varepsilon_1 S_2(x,t)$$
$$-\varepsilon_1\xi_1 D_{1i}\frac{\partial^2 C_{1i}(x,t)}{\partial x^2}+\varepsilon_1\xi_1\lambda_i C_{1i}(x,t)-\varepsilon_1\xi_1\lambda_{i-1}C_{1(i-1)}(x,t)=0 ,$$
$$x>0\,,\quad t>0 \qquad (1)$$

where ε_1 is the ratio of the liquid volume to total volume in the fracture, [-],

$C_{1i}(x,t)$ is the amount of species sorbed on the colloid per unit volume of the solid colloid, [kg/m^3],

v_{1i} is the colloid pore velocity in the fracture, [m/yr],

D_{1i} is the colloid dispersion coefficient, [m^2/yr],

λ_i is the decay constant, [1/yr],

ξ_1 is the constant volume fraction of natural colloids in the fracture liquid, [-].

Here, the concentration is assumed to be well mixed across the thin aperture of the fracture. Also the amount of the natural colloid is assumed to be so large compared with that of the pseudo-colloid that ξ_1 is set to be constant.

For the same specie as a solute in the liquid in the fracture

$$\varepsilon_1 \frac{\partial C_{2i}(x,t)}{\partial t} + \varepsilon_1 v_{2i} \frac{\partial C_{2i}(x,t)}{\partial x} - \varepsilon_1 S_2(x,t) + \varepsilon_1 S_3(x,t)$$
$$-\varepsilon_1 D_{2i} \frac{\partial^2 C_{2i}(x,t)}{\partial x^2} + \varepsilon_1 \lambda_i C_{2i}(x,t) - \varepsilon_1 \lambda_{i-1} C_{2(i-1)}(x,t) + \frac{q_i(x,t)}{b} = 0,$$
$$x > 0, \quad t > 0 \tag{2}$$

where $C_{2i}(x,t)$ is the solute concentration in the fracture liquid, [kg/m^3],

v_{2i} is the solute pore velocity, [m/yr],

D_{2i} is the solute dispersion coefficient, [m^2/yr],

and $q_i(x,t)$ is the diffusive solute flux into the rock matrix, [kg/m^2·yr], given by

$$q_i(x,t) = -\varepsilon_p D_p \left.\frac{\partial N_i(x,y,t)}{\partial y}\right|_{y=b} \tag{3}$$

where ε_p is the rock porosity, [-]

D_p is the solute diffusion coefficient in the rock matrix pores, [m^2/yr],

and $N_i(x,y,t)$ is the solute concentration in the pore water in the rock matrix, [g/m^3]:

$$R_p \frac{\partial N_i(x,y,t)}{\partial t} - D_p \frac{\partial^2 N_i(x,y,t)}{\partial y^2} + R_p \lambda_i N_i(x,y,t) - R_p \lambda_{i-1} N_{i-1}(x,y,t) = 0,$$
$$x > 0, \quad t > 0, \quad y > 0 \tag{4}$$

where R_p is the solute retardation coefficient in the rock matrix.

For solute species sorbed on the stationary fracture

$$(1-\varepsilon_1)\frac{\partial C_{3i}(x,t)}{\partial t} - \varepsilon_1 S_3(x,t) + (1-\varepsilon_1)\lambda_i C_{3i}(x,t) = 0,$$
$$x > 0, \quad t > 0 \tag{5}$$

where $C_{3i}(x,t)$ is the concentration of a sorbed solute on the stationary fracture.

For pseudo-colloids on the stationary fracture:

$$(1-\varepsilon_1)\xi_2 \frac{\partial C_{1i}(x,t)}{\partial t} - \varepsilon_1 S_1(x,t) + (1-\varepsilon_1)\xi_2 \lambda_i C_{1i}(x,t) = 0,$$
$$x > 0, \quad t > 0 \tag{6}$$

Linear sorption equilibrium between the solute species in the fracture liquid and the same species sorbed on the colloid is assumed. Both the solute species and the colloids in the fracture liquid are assumed to undergo the linear sorption equilibrium with the fracture solids:

$$K_{d1i} = \frac{\xi_2}{\xi_1}, \qquad K_{d2i} = \frac{C_{3i}(x,t)}{C_{2i}(x,t)}, \qquad K_{d3i} = \frac{C_{1i}(x,t)}{C_{2i}(x,t)} \tag{7}$$

Adding equations (1) through (6) with equations (7), the equation for $C_{1i}(x,t)$ is obtained in terms of the effective retardation factor R_i, the dispersion coefficient D_i^*, and the velocity v_i^*.

$$R_i \frac{\partial C_{1i}(x,t)}{\partial t} + v_i^* \frac{\partial C_{1i}(x,t)}{\partial x} - D_i^* \frac{\partial^2 C_{1i}(x,t)}{\partial x^2} \quad , \quad x > 0,\ t > 0 \tag{8}$$
$$+R_i \lambda_i C_{1i}(x,t) - R_{i-1}\lambda_{i-1} C_{1(i-1)}(x,t) + \frac{q_i(x,t)}{\varepsilon_1 b} = 0$$

where $R_i = \xi_1\left(1 + \frac{1-\varepsilon_1}{\varepsilon_1}K_{d1i}\right) + \left(1 + \frac{1-\varepsilon_1}{\varepsilon_1}K_{d2i}\right)\frac{1}{K_{d3i}}$ (9)

$$v_i^* = v_i\left(\xi_1 + \frac{v_{2i}}{K_{d3i} v_{1i}}\right) \tag{10}$$

$$D_i^* = D_{1i}\left(\xi_1 + \frac{D_{2i}}{D_{1i} K_{d3i}}\right) \tag{11}$$

The initial and boundary conditions are

$N_i(x,\infty,t) = 0,\quad x > 0,\quad t > 0$ (12)

$N_i(x,b,t) = \dfrac{C_{1i}(x,t)}{K_{d3i}},\quad x > 0,\quad t > 0$ (13)

$N_i(x,y,0) = 0,\quad x > 0,\quad y > 0$ (14)

$C_{1i}(0,t) = K_{d3i} f_i(t),\quad t > 0$ (15)

$C_{1i}(\infty,t) = 0,\quad t > 0$ (16)

$C_{1i}(x,0) = 0,\quad x > 0$ (17)

where $f_i(t)$ is the arbitrary time function.

The semi-analytical solution of equations (8) and (12) to (17) can be found in Ahn's analysis on the solute only migration in fractured porous media.

3. Numerical Analysis

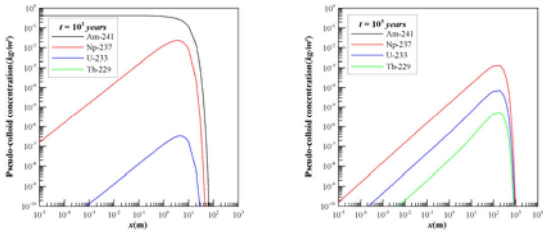

Fig. 2. Concentration profile along the fracture for a four-member decay chain.

In Fig. 2, the concentration profile of the Americium decay chain along the fracture for time ranging between 10^2 and 10^5 years are calculated. We assumed that other radionuclides are generated by the decay chain of the Americium. It can be seen from Fig. 2 that after 10^2 years the peak concentration of ^{241}Am is greater than that of other radionuclides. However, after 10^5 years, most ^{241}Am has decayed to ^{237}Np, such that maximum concentration of ^{237}Np is 10^2 times greater than that of ^{241}Am. After 10^5 years, ^{241}Am concentration decays to a negligible quantity, and other radionuclides remain in the system in a significant amount.

3. Conclusions

In this study, we use ^{241}Am→^{229}Th as the example to study the radionuclide decay chain transport in the presence of colloids. Although the decay chain may not be one of the most important cases for HLW disposal, the model can still present a useful description for further studies related to different types of four-member radionuclide decay chains along a fracture.

ACKNOWLEDGEMENT

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