Processes and Data on the Radionuclide Migration and Retardation in the Fractured Rock

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1. Introduction

In the safety assessment of radioactive waste repositories, it is necessary to understand how radionuclides migrate through a surrounding geological media. The main migration mechanism of radionuclides in the crystalline rock surrounding a waste repository would be the advective-dispersive migration with a flowing groundwater through rock fractures. Radionuclides also diffuse into the rock matrix adjacent to fractures and are retarded by the sorption on pore surfaces in the matrix depending on their sorption properties. Radionuclides with a higher sorption capacity may also be associated with natural aquatic colloids form radioactive pseudo-colloids and then fast migrate through rock fractures. Thus, in this study, we have mainly focused on the migration processes and data for the sorption and rock matrix diffusion of radionuclides in the fractured granite.

2. Rock Matrix Diffusion

Diffusion and sorption are the main retarding mechanisms in the migration of radionuclides through geological media. It is necessary to understand diffusion mechanism and to quantify the diffusion processes of radionuclides into rock matrix for the safety assessment of radioactive waste repositories. Thus, many countries have measured the diffusivities of radionuclides in the rock matrix. The apparent diffusion coefficient D_a (m^2/s) is usually adapted to represent the overall diffusion process by considering the retardation capacity of the medium and it is defined as follows:

$$D_a = \frac{D_e}{\varepsilon + K_d \cdot \rho_d} \quad (1)$$

where D_a and K_d are the effective diffusion coefficient (m^2/s) and the distribution coefficient (m^3/kg) of the radionuclide and ε and ρ_d are the porosity and dry density of the medium, respectively (kg/m^3).

The rock matrix diffusion coefficients (D_e and D_a values) of the some radionuclides in the crystalline rocks suchas granite are listed in Table I [1]. The rock matrix diffusion coefficients listed in Table I are recommended values based on the experimental data with Korean granitic rocks. As shown in Table I, the diffusivities of the actinides show values with wide ranges since the properties and chemical species of the actinides strongly depend on individual geochemical environments.

Table I: Rock Matrix Diffusion Coefficients of Radionuclides

Nuclide	D_e (m^2/s)	D_a (m^2/s)*
I	$8.3\sim230\times10^{-14}$	5×10^{-11}
Cs	8.8×10^{-14}	6×10^{-16}
Sr	$3.3\sim27\times10^{-14}$	6×10^{-14}
U	3.6×10^{-14}	3×10^{-18}
Th	6.3×10^{-15}	5×10^{-19}
Am	4×10^{-14}	5×10^{-18}
Pu	4×10^{-14}	3×10^{-18}
Cm	4×10^{-14}	5×10^{-18}
Np	$4\sim25\times10^{-14}$	3×10^{-18}
C	5×10^{-14}	2×10^{-14}
Ni	2.8×10^{-14}	5×10^{-16}
Se	4×10^{-14}	1×10^{-14}
Nb	4×10^{-14}	1×10^{-17}
Tc(IV) Tc(VII)O_4^-	4×10^{-14}	1×10^{-17} 8×10^{-12}
Sm	4×10^{-14}	7×10^{-18}

* D_a values are representative values

3. Sorption onto Granite Rock

The equilibrium partition constant usually called a distribution coefficient (K_d) has been used to quantify a sorption that is kinetically fast and reversible. However, the application of K_d in the performance assessment of a radioactive waste repository has been limited since this value is only valid for the conditions studied. Many factors such as the amount and type of minerals, the composition of the aqueous phase, pH and Eh play important but widely different roles in the sorption [2]. Thus the development of a methodology for the selection of a suitable K_d from a vast range of experimentally determined K_d values is pivotal for the safety assessment of radioactive waste repositories.

The NEA Sorption Database (SDB) was developed to provide comprehensive and complete sorption data for the performance assessment for a radioactive waste repository [2]. The use of the SDB, however, has been limited because of the complexity of its manipulation and the absence of an analysis tool for the selection of suitable sorption data. KAERI (Korea Atomic Energy Research Institute) has established a sorption database for the performance of a radioactive waste disposal. The software developed in KAERI, SDB-21C, is a graphic user interface (GUI) program that provides efficient and user friendly tools for evaluating a large amount of sorption data. The database of K_d compiled in the program contains about 11,000 NEA data and more than 4,000 KAERI data.

Table II shows the distribution coefficient K_d values obtained from domestic experimental studies for the domestic granite and groundwater [3]. The granite used in the sorption experiments was taken from a declined borehole at a depth of 157 m in KURT (KAERI Underground Research Tunnel). The granite mainly consists of quarts (32.13%), plagioclase (47.07%), biotite (7.2%), microcline (6.8%), chlorite (6.4%), and muscovite (0.4%). The groundwater was sampled from YS-01 borehole located witin KAERI research area, Daejeon (pH=9.94, Eh=-161mV).

Table II: Distribution Coefficients of Radionuclides

Nuclide	K_d (m^3/kg)	K_d^* (m^3/kg)
Ni	1.706 – 5.667	1
Se	0 – 0.0024	0
Zr	0.028 – 1.077	0.2
Pd	0.030 – 2.965	0.03
Sn	0.021 – 0.337	0.02
Cs	0.052 – 0.073	0.05
Sm	0.748 – 15.139	1
Th	4 – 7.4	5
U	0.0032 – 0.0045	0.005

K_d^* are representative K_d values

3. Sorption onto Colloids

If radionuclides become attached to mobile colloidal particles to form pseudo-colloids, the transport behaviors of these radionuclides would be different from the dissolved radionuclides. To assess the importance of the mobile colloids on the radionuclide migration at a particulate disposal site, information is required on both the total mobile load of the radionuclide and colloids as well as the distribution of the radionuclide between the colloidal and dissolved phases. Sorption properties of radionuclides onto bentonite colloids and natural colloids can provide information about the colloid migration and sorption in geological media.

Generally, the sorption coefficient of radionuclide for colloid is called a pseudo-colloid formation constant (K_{pc}) and given as follows:

$$K_{pc} = \frac{PC}{C \cdot C_c} \qquad (2)$$

where C is the concentration of the radionuclide in the solution (mol/L), C_c is the concentration of the colloids in the solution (kg/L), PC is the concentration of the radionuclide sorbed on the colloids per unit volume of the solution (mol/L), K_{pc} is the equilibrium pseudo-colloid formation constant (L/kg).

Pseudo-colloid formation constants of some radionuclides such as Sr, Am, and U for natural groundwater colloids and artificial silica and bentonite colloids are shown in Table III [1]. Many experimental results showed that association of strongly sorbing radionuclides with colloids could have a significant influence on their migration processes if the colloid concentration is sufficiently high and the colloids are mobile [4]. Thus recently the pseudo-colloid formation of radionuclides with colloids has been given the highest priority because pseudo-colloids formed in the geosphere could migrate faster than the dissolved radionuclides.

Table III: Pseudo-Colloid Formation Constants

Nuclide	Colloid	Solution	K_{pc} (mL/g)
Am	Natural colloids	Granite groundwater	7×10^4 ~ 7×10^5
Sr	Natural colloids	Granite groundwater	3×10^2 ~ 1×10^3
U	Silica fumed	pH 4~10 Carbonate	10^4 ~10^5
U	Ca-bentonite colloids	$NaClO_4$	10^4 ~ 10^7

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