Diffusion Properties of Radionuclides on Altered Granitic Rock under Aerobic Condition

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1. Introduction

As a method for the permanent disposal of high-level radioactive waste, a multi-barrier system consisting of a disposal container, buffer material, backfill, and natural rock is being considered and studied as the most effective method. Among them, it is necessary to evaluate the migration characteristics of nuclides in the rock as part of the natural barriers. The retardation mechanism of nuclide migration in the deep environment can be broadly divided into sorption and diffusion. In this study, the diffusion characteristics of nuclides in rocks based on oxidizing conditions of the Korea Atomic Energy Research Institute's Underground Research Tunnel (KAERI Underground Research Tunnel, KURT) were collected, analyzed, and evaluated for the purpose of safety assessment for the deep geological disposal system of high-level radioactive waste.

2. Methods and Results

2.1 Materials

Experiments on the diffusion of nuclides were conducted using altered granite near the KURT site. Samples of altered granite were processed to prepare test coupons for diffusion experiments. The mineralogical characteristics of the altered granitic rock coupons were confirmed using X-ray diffraction spectroscopy (XRD) and polarizing microscopy, while physicochemical properties such as porosity, density, and elemental composition were determined. Synthetic groundwater was prepared with reference to the groundwater composition within KURT, and a through-diffusion device equipped with rock coupon was fabricated and used for the diffusion experiments (Fig. 1).

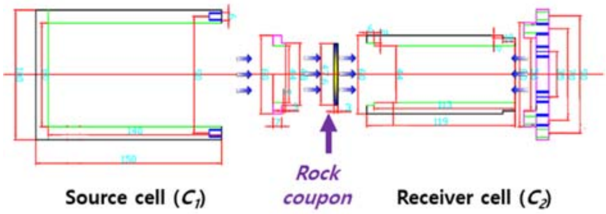

Fig. 1. Apparatus for through diffusion experiment.

2.2 Calculation of apparent diffusion coefficients

To reach steady-state conditions, it takes a long period for nuclides to pass through porous rock coupon. Once the experimental system reaches steady state, linear regression analysis can be conducted on the concentration values of the solute over time. Through this process, the apparent diffusion coefficient (D_a) of the solute through the rock was derived using the equations below:

$$\frac{C_2(t)}{C_1} = \frac{D_a R}{lh} t - \frac{lR}{6h} \quad (1)$$

$$R = \varepsilon + (1 - \varepsilon)\rho K_d \quad (2)$$

where, C_1 [μg/L]is concentration in the source cell, $C_2(t)$ [μg/L] is the concentration in the diffused cell at time t [s], R is the rock capacity (retardation) factor defined as the ratio of the overall concentration of diffusant in the rock coupon to the concentration in the solution accessible for diffusion, l [m] is thickness of the rock coupon, h [m] is length of the diffusion cell, ε porosity of the rock coupon, ρ is density of the rock coupon, and K_d [m^3/kg] is distribution coefficient.

2.3 Selection of representative radionuclides

Cesium (Cs) was selected as the representative nuclide for alkali metals, strontium (Sr) for alkaline earth metals, technetium (Tc) for transition metals, and chloride (Cl) and iodine (I) for anionic nuclides. The D_a values for each nuclide through the rock were obtained through through-diffusion experiments. The experimentally determined D_a values were compared with those proposed in the technical reports of the Swedish Nuclear Fuel and Waste Management Company (SKB) [1] and various research literature [2-6]. Recommended D_a values for use in safety assessments were proposed based on this comparison.

2.4 Characteristics of rock coupon

The mineral composition of the rock coupon used in this study was confirmed by XRD analysis to consist of albite (31.9 wt.%), quartz (34.2 wt.%), microcline (12.5 wt.%), muscovite (17.8 wt.%), calcite (2.5 wt.%), and chlorite (1.3 wt.%). Through observation using a polarizing microscope, it was confirmed that the rock coupon consists of altered granite, partially transformed into biotite through past metamorphic processes. Table I shows average values of thickness, density, and porosity of the rock coupon.

Table I: Physical properties of rock coupon

Tickness (mm)	Density (g/cm^3)	Porosity (%)
3.02	2.60	0.22

2.5 Diffusion properties of representative radionuclides

We investigated the diffusion characteristics of radionuclides in the rock matrix by comparing the D_a value of representative radionuclides with K_d value. It was observed that as radionuclides exhibited better sorption onto the rock matrix, there was a tendency for the D_a values to decrease (Table II). Anionic radionuclides (Cl, I) exhibited higher D_a values than cationic radionuclides, suggesting faster migration. We proposed recommended D_a values for safety assessment by comparing the experimentally obtained D_a values of representative radionuclides with those from SKB's technical report and research literature. Furthermore, we proposed conservative D_a values by comprehensively reviewing data from actual field samples, such as those obtained from the Grimsel Test Site (Nagra, Switzerland), and considering influencing factors such as ion strength and matrix characteristics.

Table II: Proposed D_a value for each radionuclide

	Proposed D_a value for altered granitic rock with K_d		Reference
	K_d (mL/g)	D_a (m^2/s)	D_a (m^2/s)
Cs	3.3	1.0×10^{-12}	6.5×10^{-13} [2]
Sr	8.3	1.0×10^{-13}	2.7×10^{-14} [3]
Tc	0.2	4.0×10^{-11}	2.0×10^{-14} [4]
Cl	4.6	3.0×10^{-11}	4.0×10^{-11} [5]
I	2.9	3.0×10^{-11}	2.0×10^{-11} [6]

3. Conclusions

The diffusion of nuclides is influenced by the geochemical properties of the rock and pore water and the reaction characteristics of the nuclides. Based on data from SKB's technical report and research literature were reviewed, organized, and analyzed, and recommended D_a values were proposed for use in safety assessment. The results of this study can be used for the evaluation of migration and retardation of radionuclides in order to apply a safety assessment of a deep geological disposal system. In future research, it is necessary to extensively derive D_a values for each radionuclide in rocks under various repository environment conditions.

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