Interactions of Multivalent Nuclides through a Fractured Granite under Reducing Alkaline Conditions

Park C.K. and Cho W.J.
Korea Atomic Energy Research Institute, Daejon, Korea
ckpark@kaeri.re.kr

An experimental study has been carried out for some multivalent nuclides in a naturally fractured granite in order to understand their retarding properties under reducing alkaline conditions. The nuclides investigated in these tests were *U, Th,* and *Eu*. By adding a certain amount of hydrazine, reducing alkaline conditions were made in the experimental setup. The interaction characteristics of the nuclides have been observed by changing the Eh and pH values of the solutions. The experimental conditions are arranged in Table 1.

A fundamental concern in safety assessments of radioactive waste disposal is the potential release of nuclides to the accessible environment as dissolved constituents in groundwater. The important mechanism of retarding is sorption and diffusion onto minerals present along groundwater flow paths. Thus, a quantitative understanding of nuclide sorption behavior is important in evaluating the suitability of proposed repository sites. However, this understanding is complicated by the possible dependence of sorption processes on various geochemical parameters, including aqueous solution properties such as pH, Eh, temperature, ionic strength, nuclide concentration, and complexing ligands as well as sorptive phase characteristics such as composition, surface area, sorption site density and surface charge. The dependence of sorption on various parameters makes it difficult to describe and predict nuclide retardation and transport in geochemical systems of variable and composite mineralogical composition and changing aqueous speciation. In this study, we are focused on the effects of reducing and alkaline conditions in a fractured system.

The fractured rock was sampled in a domestic quarry. This rock has an interconnected porosity of 0.37 % with the specific gravity of 2.55. Before the migration experiment, the granite block have been sink in the water for more than 3 months to make the block saturated with water.

Table 1 experimental conditions and concentrations of the nuclides

	Solution					Initial Conc. (ppm) of		
	Eh(mV)	pH	DO(mg/l)	HCO_3	CO_3 (mg/l)	U	Th	Eu
Exp.1	- 400±100	8.9±0.3	< 1.5	< 1900	< 610	12	9	5.5
Exp.2	+ 200±50	8.2±0.2	< 2.8	< 35.1	-	278	251	127
Exp.3	+ 400±50	7.3±0.2	2.6	-	-	2374	459	880

Fig.1 shows the experimental setup with a block of fractured natural granite with dimensions of 100x60x40 (cm). Nine boreholes were drilled in the upper block, orthogonal to and ending at the fracture surface. Before the migration test, the rock blocks are submerged in the water container to be saturated with water. The water in the container has kept almost a constant temperature of 20^0C. Through the nine boreholes hydraulic test was performed to estimate the transmissivity and aperture distribution. After completing the hydraulic characterization, GM tubes were placed in the seven boreholes to measure the radioactivity of the radionuclide migrating through the fracture. The signal of the detectors can be stored in the data acquisition system.

The aperture distribution in the fracture was characterized by hydraulic tests with a variable aperture channel model. Transport processes of nuclides were simulated with a particle tracking method. By comparing the simulated results to the experimental elution curves, it was checked not only the effects of matrix diffusion and geochemical conditions, but also the validity of static sorption data from the viewpoint of retardation.

The experimental results are shown in Figures 2, 3, & 4. A large portion of uranium was sorbed little on the fracture surface and transported with the nonsorbing tracer under aerobic condition. Europium were sorbed strongly on the rock surface and did not eluted out. Very small portion of thorium moved faster than the expected from the batch data, and it seemed that some portion of thorium moved in the form of chemical complex, colloid, without sorption. Generally the retardation factors obtained from the migration tests showed lower values than those obtained from the static sorption data due to sorption kinetics and contact time.

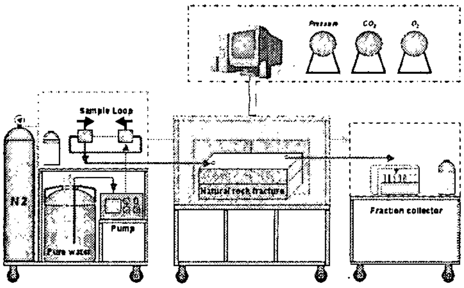

Fig.1. Experimental setup

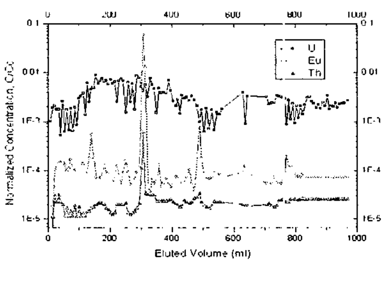

Fig.2. Elution curves U, Th & Eu under reducing conditions

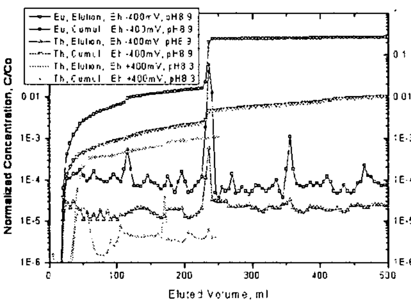

Fig.3. Elution of Th & Eu under various condt.

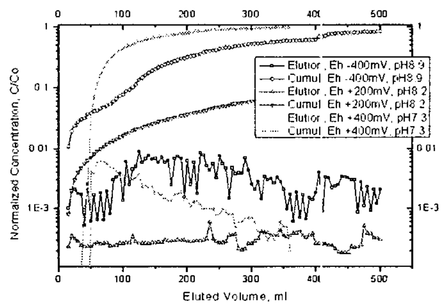

Fig.4. Elution and cumulative curves of U