Transport Properties of U, Th, Eu through a single Rock Fracture under Aerobic Conditions

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1. Introduction

A disposal research program for HLW has been carried out since 1997 with the aim of establishing the preliminary concept of geological disposal in Korea. The migration in the rock fracture has been studied in development of migration models [1,2] as well as lab. scale migration experiments [3]. 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. An important mechanism or retarding nuclide migration is sorption and diffusion onto minerals present along groundwater flow paths. However, this understanding is complicated by the possible dependence of sorption processes on various geochemical parameters. Thus we investigated the migration characteristics of redox sensitive nuclides such as U, Th & Eu in a rock fracture under aerobic environments.

2. Migration Model

In order to describe the fluid flow in the fractured rock, we are going to develop a generic model for describing the migration of tracers in heterogeneous flow fields. We characterized the value at each point with the hydraulic borehole test in the natural fracture. The fracture plane may be subdivided into imaginary subsquares. The fluid flow through the fracture was then calculated for a constant injection/elution rate as well as for constant pressure conditions. For a constant laminar flow, the volumetric flow rate, Q_{ij}, through the subsquares enclosed by the grid lines may be written as [4]:

$$\sum_j Q_{ij} = \sum_j C_{ij}(P_i - P_j) = E_i \qquad (1)$$

where P_i is the pressure at node *i*, Node *i* implies an index of the ith subsquare in the fracture surface. C_{ij} is the flow conductance between nodes *i* and *j* . E_i is the injection rate or withdrawal rate at node *i*. The subscript *j* stands for the four facing nodes of surrounding subsquares to node *i*. By rearranging the above equation for each node, we can obtain a system of linear equations in the form

$$[B]\ [P] = [E] \qquad (2)$$

where ***[B]*** is a coefficient matrix describing the flow conductance. The matrix ***[P]*** is an array describing the pressure distribution and ***[E]*** is an array describing net flow rates. Except for the nodes at the boundaries, the pressure at each node can be solved with an iteration method. The flow between adjacent nodes can be calculated using equation (1). After obtaining flow vectors at all nodes, solute transport can be simulated in this flow field.

A two-dimensional random-walk particle tracking algorithm is used to simulate the solute transport through the flow fields [5]. Particle displacements in each time step consisted of an advective displacement based on local velocities calculated using the pressure field and random diffusive displacement. Particles, which are representing the mass of a solute contained in a defined volume of fluid, move through a fracture with two types of motion. One motion is with the mean flow along stream lines and the other is random motion, governed by scaled probability. At the inlet, a certain amount of particles were introduced and distributed at each node between flow channels with a probability proportional to the flow rates[6]. Particles are then advected by discrete steps from node to node until they reach the outlet node at which point the arrival time is recorded.

3. Nuclide Migration Experiment

Migration experiments with sorbing tracers were being conducted with a natural fracture of 1 m scale. This rock has an interconnected porosity of 0.37 % with the specific gravity of 2.55. **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. Through the boreholes hydraulic test was performed to estimate the transmissivity and aperture distribution. GM tubes were placed in the boreholes to measure the radioactivity of the radionuclide migrating through the fracture. Two boreholes on the upper plate are selected as inlet and outlet for transporting of fluid. Various kinds of chemical species were used ; *THO*, *Cl*, *Eosine*, *Sr*, *Cs*, as well as *U*, *Th*, *Eu* as redox sensitive tracers. 1.2 *ml* aliquots of solution containing tracers were injected as a band input function into the inlet borehole in separate campaigns, fed with a HPLC pump through the fracture at a flow rate of 0.5 *ml/min* and collected at the outlet borehole. The eluted solution was collected using a fraction collector.

4. Study Results

Fig.2 shows the simulated flow field based on the hydraulic test. The flow field is well developed and no dead zone. The mean aperture is about 0.1 *mm*. The pressure distribution shows symmetric along the diagonal line between the inlet and outlet. To analyze the migration characteristics of tracers according to their chemical properties, elution curves and cummulative elution curves were examined in **Fig.3.**

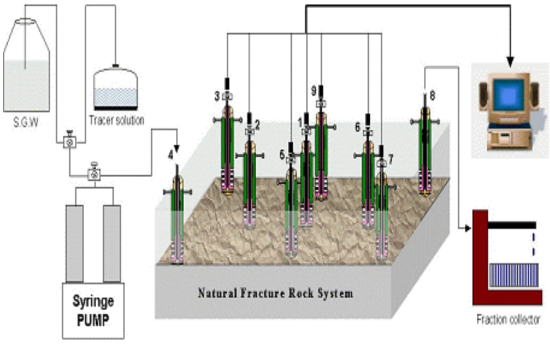

Figure 1. Schematic diagram of the experimental setup

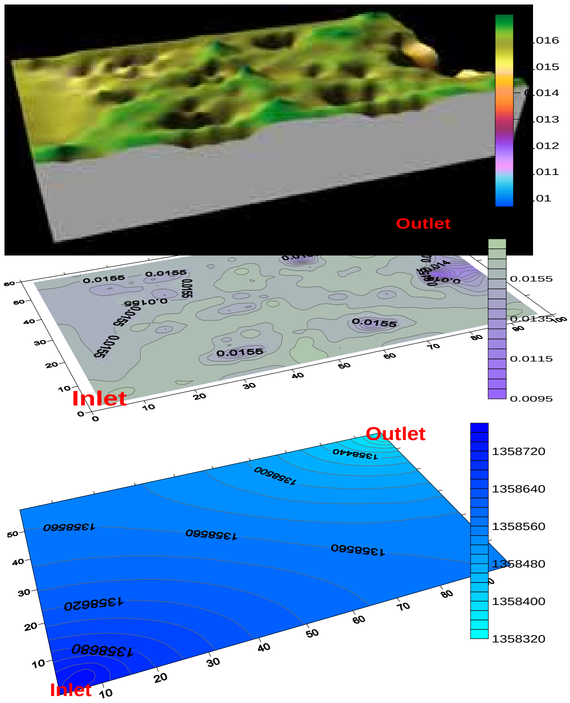

Fig.2. Simulated Flow field

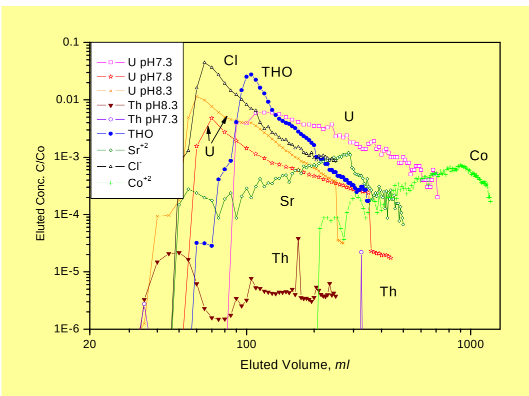

Fig.3. Elution curves of the tracers through the fracture

The anions and polymeric organic dyes migrate faster than the tritium in the natural fracture and they are recovered almost 100% at the exit. While tritium was recovered only 83%. Because the surface of the rock fracture is negatively charged, the anion could be expelled by the rock surface and thus can not access to the pore by the anion exclusion effect. Therefore, it could be concluded that tritium diffuses into the rock pores, but anions and polymeric tracers hardly diffuse into the rock matrix.

The migrating sorbing tracer interacts with the fracture surface and it retards as much as their sorption capacity. The degree of retardation is usually expressed as the retardation factor, *R.* The *R* values from the elution curve was smaller than those from sorption data. It seems that the sorbing tracers did not interact with the rock surface in the flow rate of 0.2 ml/min. A large portion of uranium sorbed little on the fracture surface and transported with the nonsorbing tracers. Cesium and europium sorbed strongly on the surface and did not eluted out. A little portion of Th makes complexes like colloids and migrate without sorption.

5. Concluding Remarks

The developed variable aperture channel model was successfully applied in describing the migration of radionuclides in the rock fracture. Results from the modelling of the transport show that diffusion into the interconnected micropore space in the rock mass has an effect on the retardation, especially for the case of tritium. The redox sensitive nuclides such as *U, Th, & Eu* moved faster than the expected from the batch sorption data. Thus, it is needed to study geochemical sensitivity on the actinides and lantanides.

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