The Effect of Fractured Rock to Radionuclides Transport Through Unsaturated Zone

Won-Seok Kim*, Sangsoo Han, and Wooyong Um

Pohang University of Science and Technology (POSTECH), ChungAm-Ro 77, Pohang, South Korea

*wkim23@postech.ac.kr

1. Introduction

The potential release of radionuclides from low and intermediate-level radioactive waste (LILW) may pose environmental problems to adjacent areas. In addition, the presence of released radionuclides can affect human health [1]. Released radionuclides can be transported by the flow of rainfall, exist with pore water through fractures in unsaturated zone, and is reached to groundwater flow. Released radionuclides to vadose zone and ultimately groundwater can be transported by groundwater flow through porous materials or fractures [2]. It is important to investigate the effect of fractured rock to radionuclides transport through vadose zone. In this study, batch adsorption, batch diffusion, and flow-through column experiments were conducted to understand the sorption and transport behavior of radionuclides under fractured rock in unsaturated zone, and the safety assessment and long-term performance of repository.

2. Materials and methods

2.1 General information

To investigate the relationship between the sorption of radionuclide and site characteristics which fractured rock in vadose zone, batch sorption, batch diffusion, and flow-through column experiment for radionuclides such as ^{3}H, ^{99}Tc, and ^{90}Sr were conducted using groundwater and solid materials from the LILW repository site. Fractured rock samples were collected in the LILW repository site, and it was characterized by X-ray microtomography (XMT) analysis and fractured filling/coating material was investigated by X-ray diffraction (XRD).

2.2 Experiments

Solid material of fractured rock sample (1.0 g for ^{3}H, 1.0 g for ^{99}Tc, 0.5 g for ^{90}Sr) was used in 10 mL of groundwater for the batch sorption experiments. Specimen sample (2 × 2 × 0.5 cm) was used for batch diffusion experiments. Column sample (5.0 × 12.5cm) was used for flow-through column. In order to compare the radionuclide adsorption to fracture filling/coating material, experiments were done with and without fracture filling/coating materials. The size of solid material is 75-250 µm.

Solid material was pre-equilibrated through contact with a sufficient amount of groundwater for one day for the sorption experiments. Pre-equilibrated step was repeated until the pH of the solution was maintain with the pH of groundwater (pH=6.8). After pre-equilibration, control batch test which without radionuclide was performed to determine the initial concentrations of the radionuclides and the mass loss by sorption onto bottle walls. The radionuclide was spiked into the pre-equilibrated reacting bottles to attain the required radionuclide concentration (^{3}H: 100 Bq/mL, ^{99}Tc: 100 Bq/mL, ^{90}Sr: 20 Bq/mL). Solid material was shaken at 80 rpm with radionuclide spiked groundwater for 14 days. The pH was adjusted to maintain the initial pH of the groundwater by 0.1M HNO_3 or NaOH solution during experiment. After finish the batch sorption experiment, the solution was separated by using a syringe filter of 0.45 µm pore size (Whatman) and stored to determine the concentration of radionuclides. All batch sorption experiments were carried out in triplicate.

The diffusion of radionuclide were investigated using diffusion reactors which shown in Fig. 1, and it was divided by two part. Tracer and radionuclide was inserted to one part and it was transported to the other part by diffusion through specimen which installed in middle of reactor.

The fractured rock sample was setting inside of column and the condition of column was controlled in unsaturated (water content: 35%) by syringe pump. The flow of groundwater with radionuclide was controlled by syringe pump.

(a) (b) (c)

Fig. 1. Experiment for (a) batch sorption, (b) diffusion, and (c) flow-through column.

3. Results and discussion

The XMT and XRD results show the characterization of fractured rock sample. The distribution coefficient of radionuclide was calculated after experiment and compared with and without fracture filling/coating material.

3.1 XMT and XRD

XMT, a nondestructive three-dimensional imaging technique, was applied to demonstrate its capability to visualize the mineralogical alteration and microstructure changes in rock sample. Fig. 2 shows the XMT images of fractured rock surface. XRD result (Fig. 3) shows that heulandite and laumontite was major component of fracture filling/coating material.

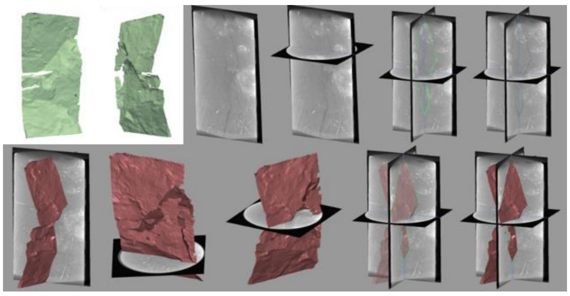

Fig. 2. X-ray microtomography image of fractured rock surface.

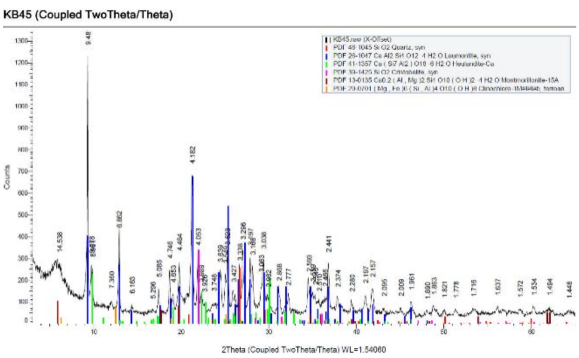

Fig. 3. X-ray diffraction results of fractured rock.

3.2 Radionuclide transport behavior

The fracture filling/coating materials were significantly affect to the distribution coefficient of radionuclide sorption. The distribution coefficient of ^{90}Sr (45.1 mL/g) with fracture filling/coating materials is higher than without fracture filling/coating materials (30.3 mL/g). In case of ^{99}Tc, there are similar distribution coefficients in both with and without fracture filling/coating materials (0.9 and 1.1 mL/g). The batch diffusion and flow-through column experiments were also conducted using the same fractured rock sample, and the results of diffusion and column experiments showed similar trend of radionuclide sorption and transport to sorption experiment.

4. Conclusion

When the fracture filling/coating materials are exist, distribution coefficient of radionuclide is higher than without fracture filling/coating materials. This study involved a geochemical characterization of the LILW repositroy, and was performed as a part of the site characterization effort. The results of this study provide more basic information and insight for the safety assessment of LILW repository.

REFERENCES

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