Evaluation of Nuclide Behavior Due to Non-Reactive Gas Breakthrough Buffer Material

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1. Introduction

High-level radioactive waste disposal is a significant challenge for countries engaged in nuclear power generation. To address this issue, deep geological disposal has been recommended as a method. This approach involves constructing disposal tunnels in stable geological formations hundreds of meters underground and using metal canisters and bentonite buffer materials to isolate high-level radioactive waste for long periods. The metal canister and buffer are referred to as engineered barriers, which prevent the external leakage of radionuclide and ensure their long-term isolation. The rock in which the disposal tunnels are constructed acts as a natural barrier, preventing the entry of radionuclide into the ecosystem and their migration into human habitats. The disposal system, including both the engineered and natural barriers, is termed a multi-barrier system. Through this system, high-level radioactive waste can be safely disposed of [1].

If the metal canister is damaged, groundwater can infiltrate the canister, potentially leading to the external leakage of radionuclide. The leaked radionuclide may diffuse through the buffer material and reach the natural barrier. However, the groundwater flow rate in the deep underground is very slow, which limits the diffusion of radionuclide.

In the disposal environment, there are mediators that can overcome this slow flow rate, notably colloids and gas. Colloids can move acceleratedly due to the electrostatic repulsion by the charged surfaces within the fracture zones of the rock, facilitated by their inherent charge. Additionally, radionuclide can attach to the colloids formed in the deep disposal environment and move with them. These colloids can travel faster than the average groundwater flow rate through the buffer and backfill materials to the natural barrier, thus accelerating the movement of radionuclide.

When gas accumulates at the boundary of the metal canister and the buffer material, it can break through the buffer, expelling water dissolved with radionuclide. The gas breaking through the buffer can form bubbles depending on the differential pressure, which move upwards due to buoyancy. The movement speed of these bubbles is faster than the average groundwater flow rate, and the surfaces of these bubbles can carry attached radionuclide colloids or pseudo-colloids, thus potentially accelerating the upward movement of radionuclide faster than the average groundwater flow rate.

Previous studies have conducted gas permeation experiments on buffer materials [2] and particle transport modeling by bubbles [3], but there has been no research on the transport of nuclide by particles formed as gas breaks through the buffer material. In this study, experiments were conducted to simulate the acceleration of nuclide transport behavior due to the erosion phenomenon occurring as gas breaks through the buffer material, leading to the formation of fine particles and the transport of nuclide by these particles.

2. Experimental

In this experiment, an environment simulating the deep geological disposal condition (underground at 500 m pressure conditions) was created to evaluate the behavior of nuclide caused by gas within the compressed buffer material. Throughout the experimental process, the temperature inside the pressure vessel was measured to maintain constant temperature conditions, and pressure was also measured to consistently replicate the hydrostatic condition at 500 m underground. A system was designed to automatically log the measured temperature and pressure data for analysis. Figure 1 illustrates the experimental equipment along with the remote-control system capable of logging temperature and pressure, and a camera system that allows for the long-term recording and storage of the internal phenomena.

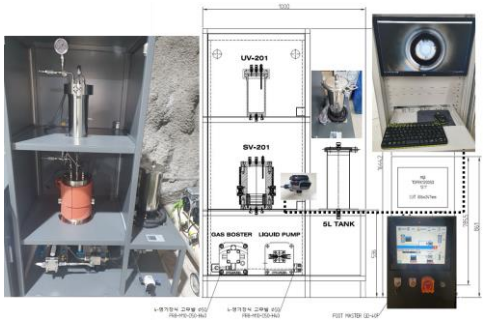

Fig. 1. Monitoring system capable of recording experimental videos in the KURT environment

3. Results and discussions

Figure 2 presents the results of particle size distribution analysis conducted after filtering the solutions from the lower reactor and the upper reactor and acquiring images through SEM analysis. The SEM images used for particle size distribution analysis indicate that the average particle size in the lower reactor is relatively large, with a significant number of particles under 500 nm in the upper reactor. It is inferred that most particles generated during the intermittent gas breakthrough process remained within the lower reactor, with their average size existing between approximately 500 ~ 1000 nm. It is assumed that smaller particles, around 250 nm in size, ascended due to buoyancy caused by bubbles and were captured within the upper reactor.

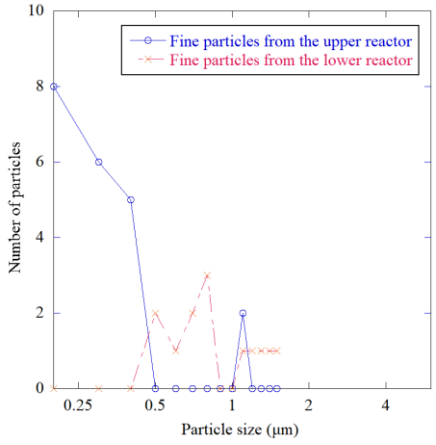

Fig. 2. Particle size distribution from SEM results of bentonite erosion particles formed during the intermittent gas breakthrough process.

The recovered solutions were analyzed using ICP-MS (GreenBio Center, Bruker aurora M90), focusing on the concentrations of cesium and iodine within the solution. For cesium, 0.425 mg was detected in the lower reactor solution and 2.771 mg in the upper reactor solution. When converted to the percentage of the introduced 133 mg, 0.32% and 2.08% of cesium moved to each reactor's solution, respectively. Iodine was detected at 42.568 mg in the lower reactor and 13.447 mg in the upper reactor, converting to 32.74% and 10.34% movement to each reactor, respectively, compared to the introduced 130 mg. This indicates that, notably for iodine, due to its high mobility characteristic of anionic nuclide, a significant amount moved upwards without being adsorbed by the bentonite. Moreover, it appears that no separate transport occurred during the bubble ascension due to buoyancy, only showing a dilution effect due to the concentration difference in the upper and lower solutions. Conversely, it seems that most of the cesium was adsorbed by the bentonite and did not move to the upper solution, with a higher concentration in the upper reactor than in the lower reactor (Figure 3). To verify the hypothesis that the higher concentration of cesium in the upper reactor was due to its adsorption onto eroded fine particles moving with the bubbles, SEM-EDS analysis was conducted to check for the adsorption of nuclide within the fine particles. The results confirmed that neither cesium nor iodine was adsorbed. The indirect evidence from these analyses suggests that relatively small bentonite particles moved upwards due to the bubbles, and during this process, cesium moved along and re-desorbed while remaining inside the upper reactor, thereby increasing the cesium concentration in the solution. Furthermore, these results are consistent with the modeling outcomes of Neretnieks et al. [3].

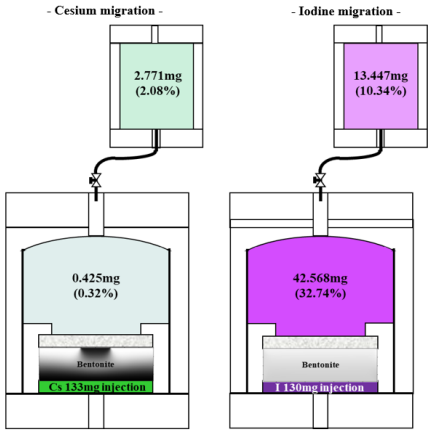

Fig. 3. Nuclide diffusion in compressed bentonite and transport by fine particles due to intermittent gas breakthrough.

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