Transport of True Colloids and Pseudo-Colloids in a Compacted Bentonite Buffer System

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***Keywords :** Deep geological repository, Colloid, Gold, Cesium, Bentonite, Transport

1. Introduction

Because of the continued generation of high-level waste (HLW), a reliable safety assessment based on the accurate modeling of radionuclide migration and retardation behavior is required for safe disposal of HLW. However, colloidal substances within the repository environment can introduce significant uncertainties in the prediction of the behavior of radionuclides. Therefore, understanding the behavior of colloidal radionuclides in the multi-barrier system of repository is critical to ensure the long-term safety of a geological disposal facility. In this context, this study investigates the transport behavior of both true colloids and pseudo-colloids through a compacted bentonite buffer system.

2. Material

2.1 High-pressure reaction cell and bentonite specimen

A high-pressure reaction cell system was installed to withstand both the swelling pressure of compacted bentonite and an injection pressure of 7 MPa. Uniform injection of the colloidal dispersion was achieved with a sintered metal filter (pore size: 0.5 μm) in combination with a porous holder. The apparatus for the reaction cell system used in this study is shown in Fig. 1.

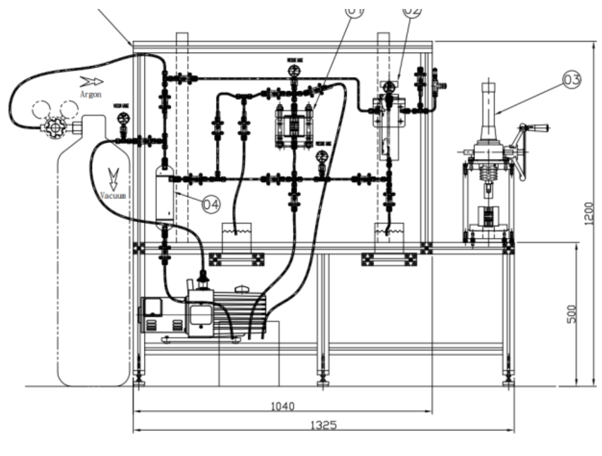

Fig. 1. High-pressure reaction cell system.

Bentonil-WRK bentonite (Clariant Korea) was compacted into cylindrical specimen with a diameter of 30 mm and a height of 20 mm, at a dry density of 1.63 ± 0.01 g/cm^3. To prevent lateral flow during the transport experiment, rubber O-rings were placed along the side of the bentonite specimen.

2.2 Colloidal gold and Cs^+-bentonite dispersions

A commercial colloidal gold dispersion (BIONEER) with a concentration of 100 mg/L was employed as the true colloid sample. The mean particle size of colloidal gold was determined to be 12 ± 2 nm by Transmission Electron Microscopy (TEM).

The pseudo-colloid was prepared by adsorbing Cs^+ ions onto colloidal bentonite. A stock solution of $[Cs^+]$ = 10^{-4} M was obtained by dissolving $CsClO_4$ (Sigma-Aldrich) in deionized water. A colloidal bentonite dispersion was synthesized by dispersing the raw bentonite powder in deionized water at a concentration of 100 g/L. The resulting suspension was subsequently centrifuged at 2,307 × g for 20 minutes, and the supernatant was collected as the stock dispersion. The pseudo-colloid was then prepared by contacting the colloidal bentonite dispersion with the Cs^+ stock solution, resulting in initial bentonite colloid and Cs^+ concentrations of 70 mg/L and 10^{-6} M, respectively. The pH was adjusted to 9.0 with a 0.1 M NaOH (Merck) solution, and the ionic strength was maintained at 0.01 M by adding 0.1 M $NaClO_4$ (Sigma-Aldrich) solution. Approximately 85% of the Cs^+ ions were adsorbed onto the colloidal bentonite, and Dynamic Light Scattering (DLS) indicated a mean hydrodynamic diameter of 537 ± 180 nm.

2.3 Analytical methods

To determine the concentrations of gold in the given system, a procedure for acid digestion was developed using aqua regia. Gold concentrations in the injection solution and the effluent were measured using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS). Additionally, TEM analysis was conducted to confirm the presence of colloidal gold particles in the effluent.

In the transport experiment involving Cs^+-bentonite pseudo-colloids, the effluent is expected to contain both free Cs^+ ions and Cs^+ ions adsorbed onto colloidal bentonite particles. Therefore, phase separation was performed with a 10 kDa centrifugal filter that had been preconditioned twice using a 0.01 M $NaClO_4$ solution. The total Cs^+ concentration in the effluent was determined from an aliquot diluted in a 4 M HNO_3 solution, whereas the filtrate was analyzed to determine the free Cs^+ ion concentration after dilution with a 2% HNO_3 solution.

3. Result and Discussion

3.1. Transport of gold true colloids

The compacted bentonite was first saturated with deionized water, and then the colloidal gold dispersion was injected at a pressure of 7 MPa over a period of 22 days. ICP-MS analysis of the effluent revealed very low gold concentrations (*ca.* 1 μg/L). A TEM image of the effluent is shown in Fig. 2, revealing spherical particles with an average size of 4.2 ± 0.9 nm. However, complementary Energy Dispersive Spectroscopy (EDS) analysis was unable to confirm the elemental composition of colloidal substances likely due to the very low concentration and the small particle size. The findings indicate that the colloidal gold particles with diameters smaller than 5 nm, contained in the injected colloidal gold dispersion, may have migrated through the pores of the compacted bentonite. It is worth noting that this size range is consistent with nanoparticles that did not undergo coarsening during the synthesis [1].

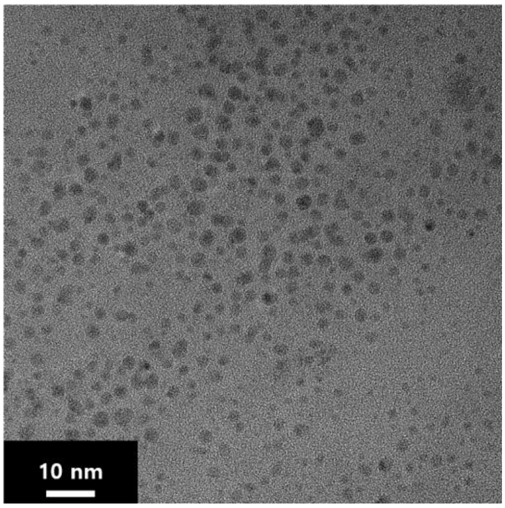

Fig. 2. Colloidal gold particles in the effluent

3.2. Transport of Cs^+-bentonite pseudo-colloids

Similar to the colloidal gold transport experiment, the compacted bentonite was first saturated with deionized water. The colloidal Cs^+-bentonite dispersion was then injected at a pressure of 7 MPa over a period of 40 days. The effluent samples were collected at 3-day intervals and both the Cs^+ concentration and effluent volume were monitored. According to the results, these parameters remained nearly constant until day 13 and then decreased sharply, as shown in Fig. 3. This trend is likely due to clogging of the pores of the compacted bentonite by the colloidal bentonite particles, which form a filter cake and impede the fluid flow [2,3].

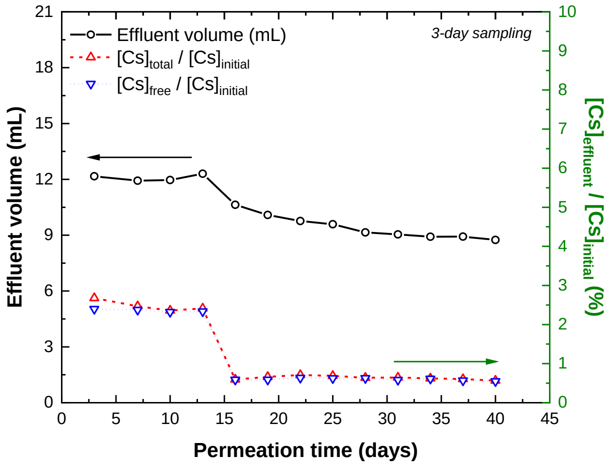

Fig. 3. Variations of effluent volume and Cs^+ concentration

4. Conclusion

In the present work, the transport behavior of colloidal gold and Cs^+-bentonite particles through the compacted bentonite buffer was investigated. The results indicate that relatively small particles, such as colloidal gold, can migrate through the compacted bentonite, although the observed gold concentration was very low. In parallel, the colloidal Cs^+-bentonite experiment revealed that these larger particles could not migrate through the pores, and instead clogged the internal pore network, which reduced the flow rate. These findings provide preliminary insights into field-scale experiments on colloid-facilitated migration of radionuclides in underground research laboratories.

Acknowledgement

This work was supported by the Institute for Korea Spent Nuclear Fuel (iKSNF) and National Research Foundation of Korea (NRF) grant funded by the Korea government [No. NRF-2021M2E1A1085202] and was supported by the 'Human Resources Program in Energy Technology' of the Korea Institute of Energy Technology Evaluation and Planning (KETEP), which was funded by the Ministry of Trade, Industry & Energy (MOTIE) of the Republic of Korea [No. RS-2024-00398425].

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