

Effect of C–S–H Colloids on Radionuclide Transport in Crushed Concrete Columns

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1. Introduction

Radioactive waste disposal facilities that employ concrete barriers must evaluate the transport behavior of radionuclides. Among the various factors, the influence of chemical additives has not yet been fully clarified. These additives can chelate radionuclides, transforming them into more mobile forms or increasing their apparent solubility [1-2]. They may also promote the colloidization of cement particles, leading to the formation of mobile colloidal phases that can act as carriers for radionuclides [3]. Such processes may significantly alter radionuclide transport behavior in cement-based barriers. Therefore, it is essential to investigate these effects.

In this study, calcium silicate hydrate (C–S–H) nanoparticles were synthesized using a polycarboxylate-based superplasticizer (PCE), a chemical additive commonly used to improve the workability of cement paste, and their effects on radionuclide transport were investigated.

2. Experimental

2.1 Materials

C–S–H colloids were prepared by a double-precipitation method. A 60 mg/L suspension of C–S–H colloids was synthesized by mixing 5 mM Na₂SiO₃ and 5 mM CaCl₂ solutions under conditions of 0.01 M NaOH and 1% PCE. For the column experiments, the suspension was diluted tenfold with artificial cement pore water (ACW).

Concrete from the Gyeongju Radioactive Waste Disposal Facility was crushed and sieved, and only particles in the 0.6–1.0 mm size range were selected. These particles were packed into empty 5 mL FPLC columns to prepare two crushed-concrete columns. The porosity was approximately 0.43, corresponding to a pore volume of about 2.2 mL.

2.2 Column transport method

All experiments were conducted in a glove box to prevent the ingress of carbon dioxide. To simulate a

radioactive waste source, a synthetic radionuclide cocktail was prepared by adding Cs, Se, Mo, U, and D₂O (water flow tracer) to ACW. Four influent solutions were prepared: (I) ACW; (II) ACW containing C–S–H colloids (0.1% PCE); (III) a multi-element cocktail containing 1% D₂O; and (IV) a solution containing both C–S–H colloids and the multi-element cocktail.

The influent was injected at a flow rate of 2 mL/h using a peristaltic pump, and the effluent was collected with a fraction collector and quantified by ICP-MS. The injection sequence for each column was as follows. Column A was operated in the order (I) → (III) → (I) → (II), whereas Column B was operated in the order (I) → (IV) → (I).

3. Results and Discussion

3.1 Characterization of C-S-H colloids

C–S–H colloids were characterized by cryo-TEM imaging and dynamic light scattering (DLS) analysis. The cryo-TEM image is shown in Fig. 1. The average particle size was measured to be 10.3 ± 5.9 nm. As shown in the DLS results in Fig. 2, the mean particle size appeared relatively large due to the presence of a small fraction of larger particles. However, the size distribution of the original suspension prior to dilution indicates that a substantial proportion of smaller particles was present.

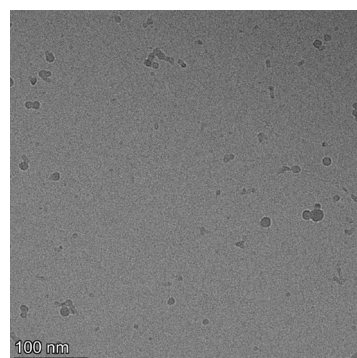


Fig. 1. Cryo-TEM image of C–S–H nanoparticles (10-fold dilution, 0.1% PCE).

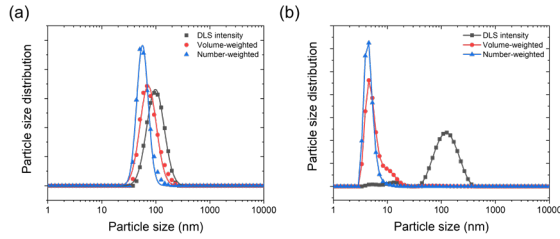


Fig. 2. Particle size distribution of C-S-H nanoparticles measured by DLS for (a) a 10-fold dilution (0.1% PCE) and (b) the original stock suspension (1% PCE).

3.2 Breakthrough curves

The concentration of the multi-element cocktail was defined as C_0 . The effluent concentration C was measured and the relative concentration (C/C_0) is presented in Fig. 3. D_2O was analyzed by Raman spectroscopy, and its breakthrough point corresponded to the flow rate of water.

Mo(VI) exhibited no retardation and migrated together with the water. Se(IV) showed only slight retardation even in the absence of colloids; in the presence of colloids, retardation was nearly negligible. U(VI) did not elute at all in the absence of colloids. However, when colloids were present, a fraction (approximately 60%) broke through, and the breakthrough coincided with the water flow. Therefore, a colloid-facilitated transport effect was clearly observed for U(VI).

Cs(I), which does not sorb onto colloids but readily sorbs onto aggregates in concrete, eluted regardless of the presence of C-S-H colloids [4]. However, in Phase III of Column A, slight desorption was observed when colloids were injected, due to partial dissolution of the concrete induced by the co-injected PCE. This effect should be attributed to PCE rather than to colloid-facilitated transport.

4. Conclusions

C-S-H colloids were synthesized and characterized under cement pore water conditions in the presence of PCE. The results indicate that C-S-H colloids can partially accelerate radionuclide transport. Radionuclides with low affinity for C-S-H, such as Mo(VI) and Cs(I), were scarcely affected, whereas U(VI), which exhibits strong selective sorption onto C-S-H, broke through rapidly and migrated with the water flow.

This study demonstrates that C-S-H colloids formed in the presence of PCE can selectively enhance the mobility of radionuclides that have a strong affinity for the colloidal phase.

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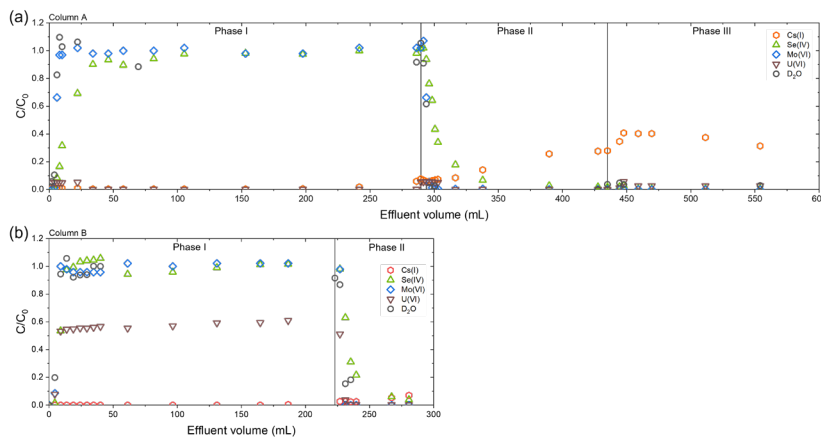


Fig. 3. Breakthrough curves for (a) Column A and (b) Column B.