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Sorption Behavior of Cesium on Calcium Silicate Hydrate under High Carbonate Ion Concentration Conditions

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Abstract

To assess radionuclide migration, Ca/Si ratio of calcium silicate hydrate (CSH) gel formed as a secondary mineral is a key factor because CSH gels effectively adsorb radionuclides, in particular cationic nuclides. However, the Ca/Si ratio of the CSH gel is changed by the leaching of Ca ions and the formation conditions. In this study, the influence of carbonate ions on the sorption of Cs ions on the CSH gel was preliminarily examined. For the sorption experiments, the concentrations of carbonate ions (NaHCO_3) were adjusted to 0 mM–1000 mM at the synthesis of the CSH gel or after a 7-day curing period. As a result, the sorption of Cs ions on the CSH gel became larger with the increase in the concentration of carbonate ions. Such sorption behavior was also observed when carbonate ions were added after curing the CSH gel. These results suggest that CSH gel formed as a secondary mineral might be able to contribute to the retardation of radionuclide migration by interacting with carbonate ions.

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1. INTRODUCTION

To more reliably assess radioactive waste disposal systems, it is necessary to understand the alteration of the natural barrier system, which is caused by using a large amount of cementitious materials during construction [1]. In particular, under the condition of high pH, a calcium silicate hydrate (CSH) gel forms with the reaction of silicic acid with Ca ions leached from cementitious materials. This CSH gel, formed as a secondary mineral in the near field of the repository, is a very important factor for the assessment because the CSH gel can contribute to the retardation of radionuclide migration via sorption effects for nuclides. For example, the authors have reported that CSH gels effectively sorb cesium (Cs) ions and barium (Ba) ions even under the condition of saline water [2]. However, natural groundwater includes many components that may affect the stability and sorption effect of CSH gels. This study focuses on the influence of carbonate ions on the stability and sorption effect of a CSH gel. In general, the concentration of carbonate ions in groundwater ranges from approximately 1 mM to 10 mM. Under high pH conditions, carbonate ions form calcium carbonate (calcite) by reacting with calcium ions. In other words, the formation of calcite causes a decrease in the Ca/Si ratio of the CSH gel due to the interaction between carbonate ions and Ca ions in the CSH gel. Such changes in the Ca/Si ratio of the CSH gel may alter its stability and sorption effect. Therefore, in this study, the sorption behavior of Cs ions on a CSH gel under the condition of coexistence with carbonate ions was preliminary examined using CSH-gel samples without a drying process [3-4] because the repository would be saturated again by the groundwater after backfilling.

2. EXPERIMENTAL PROCEDURES

Table I summarizes the conditions of the sorption experiments in this study. A CSH gel was synthesized at each Ca/Si molar ratio by mixing fumed silica (specific surface area of $300 \pm 30 \text{ m}^2/\text{g}$), CaO, and ultra-pure water in PPCO centrifuge tubes (Nalgene™, 50 mL). The initial Ca/Si molar ratios of the CSH gels were set to 0.4, 0.8, 1.2, and 1.6. The total amount of CSH gel was 1.5 g, and the liquid/solid weight ratio was set to 20 mL/g. Then, a CsCl solution was added to the samples to adjust the Cs ion concentration to 5 mM. This means that the curing and the sorption of Cs ions on the CSH gel occurred at the same time. The concentrations of the carbonate ions (NaHCO_3) were adjusted to 0 mM, 10 mM, 100 mM, and 1,000 mM. Carbonate ions were added to each tube in two ways. One was “Case (I)” wherein the carbonate ions were added in parallel to the addition of a CsCl solution as the condition of co-precipitating carbonate ions, CSH gel, and Cs ions. The other was “Case (II)” wherein carbonate ions were added after a 7-day curing period, particularly after the formation of the CSH gel. These procedures were conducted in a glove bag filled with nitrogen gas. During the curing and sorption periods, the sealed sample tubes were shaken at 120 strokes/min at 298 K. After 14 days following the synthesis of the CSH gel, the samples were centrifuged for 10 min at 3,000 rpm, and the liquid phase was filtered with a 0.45- μm membrane filter. The concentrations of Ca and Si in the liquid phase (the filtrated aliquot) were measured via Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES), and the concentration of Cs ions was measured via Atomic Absorption Spectrometry (AAS).

Table I. Conditions of the sorption experiments.

Liquid/Solid ratio [ml/g]	20 (Solution volume: 30 mL, solid weight: 1.5 g)
Ca/Si ratio [-]	0.4, 0.8, 1.2, and 1.6
Initial conc. of NaHCO_3 (carbonate ions) [mM]	0, 1, 10, 100, and 1000
Initial conc. of CsCl [mM]	5
Sorption period [days]	14
Temperature [K]	298 ± 1
Shaking rate [strokes/min]	120

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