



A new technique for removing strontium from seawater by coprecipitation with barite



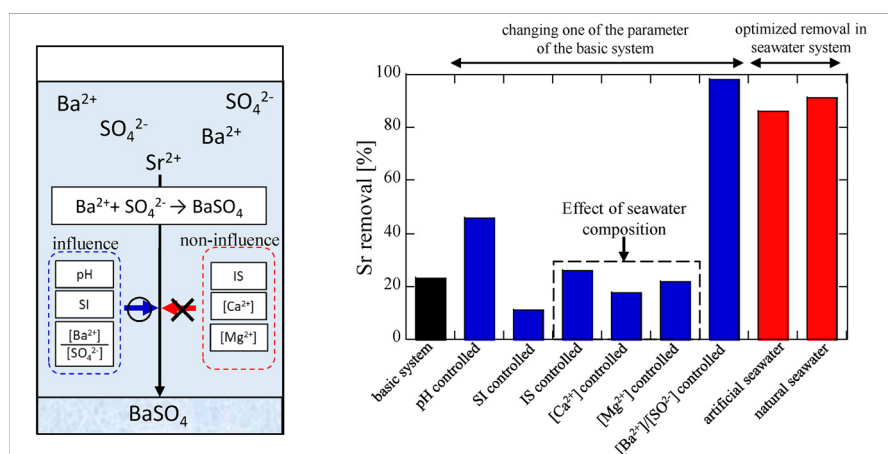
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GRAPHICAL ABSTRACT



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ABSTRACT

Strontium (Sr) removal from seawater has recently attracted attention from an environmental perspective after the Fukushima Nuclear Power Plant accident, but there is a lack of effective removal techniques for removing Sr from seawater. In the present study, we looked at the removal efficiency of Sr by using barite ($BaSO_4$) under various experimental conditions to develop techniques for the direct removal of Sr from seawater. The effects of pH, saturation state, ionic strength, competitive ions, and $[Ba^{2+}]/[SO_4^{2-}]$ ratio in the initial aqueous solution were examined. Among them, Sr uptake by barite was found to be dependent on pH, saturation state, and $[Ba^{2+}]/[SO_4^{2-}]$ ratio in initial aqueous solution, showing that most of the aqueous Sr can be removed from the aqueous solution by adjusting these parameters. However, the effects of ionic strength and competitive ions were negligible, suggesting the effectiveness of its application to removal of Sr from seawater. Batch experiments were also conducted in a seawater system, and a rather high removal efficiency of Sr from seawater (more than 90%) was achieved. Considering its high removal and retention efficiency of Sr in seawater systems, barite is a reliable material for the removal of Sr from seawater.

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1. Introduction

Strontium-90 (^{90}Sr ; half-life = 28 years) is a fission product of uranium, which is released to the environment from nuclear plant accidents [1,2]. It is known that ^{90}Sr is one of the most abundant radioactive pollutants in aqueous effluents, and the separation and removal of radio Sr ions from aqueous solutions need special attention owing to its long lifetime, high solubility, and biological toxicity of radioactive Sr. Several techniques can be used to reduce the Sr level in polluted solution such as ion exchange, chelation, adsorption, coprecipitation, and bioremediation [3–13]. Previous studies have shown the high efficiency of Sr removal by using adsorption methods. However, retention by adsorption would be unstable over the long term because changes in the surrounding physico-chemical environment would induce release of the adsorbed ions back to water [14,15].

Therefore, in the present study, we focus on the immobilization of trace elements within minerals during crystal growth, also known as coprecipitation to develop a technique for effective and stable removal of Sr from aqueous solutions. The advantages of the coprecipitation method are simplicity, short treatment time, low cost, and preservation of substituent ions in the crystal lattice for a long time, which makes it work as an engineered barrier for the retention of various ions, including Sr [12–23]. In this study, we designed and characterized methods using barite (BaSO_4) and analyzed the removal efficiency of Sr from aqueous solution by using the developed methods. Barite is a cheap and effective material and can be used to remove toxic and/or radioactive elements from polluted solutions. As a sequestering phase, barite shows following characteristics: (i) extremely low solubility [24] (ca $K_{\text{sp}} = 10^{-9.98}$ at 25 °C, 1 atm); (ii) incorporation of numerous elements because of the large ionic radii of substituted ions [15,25] (Ba^{2+} : 1.68 Å; SO_4^{2-} : 1.48 Å); (iii) high density compared to other minerals (4.5 g/cm³), which is an advantage for rapid sedimentation during coprecipitation process; and (iv) high crystal stability under wide ranges of pH, redox potential, temperature, and pressure conditions [26–35]. Thus, barite serves as a sequestering phase for the removal of toxic and/or radioactive elements from polluted solutions. For example, barite acts as an ideal host mineral for Ra^{2+} in fresh or brine water because of its high stability and higher incorporation of Ra^{2+} relative to other minerals [27–30].

In the present work, we studied the coprecipitation mechanism of barite for developing a technique for the direct removal of Sr from aqueous solutions instead of precipitation of pure solid. In the latter case, the solubility of BaSO_4 ($K_{\text{sp}} = 10^{-9.97}$) is significantly lower than that of SrSO_4 ($K_{\text{sp}} = 10^{-6.62}$); thus Sr coprecipitated with barite can be more recovered readily from an aqueous solution than the precipitation of pure SrSO_4 solid [23]. In addition, barite is resistant to dissolution and is very stable in the crystalline form from the viewpoint of the immobilization of Sr, suggesting high applicability of the barite coprecipitation method. Coprecipitation is a structural incorporation process during crystal growth, and the precipitates do not release the immobilized ions unless the host mineral is dissolved.

This study examined the removal efficiency of Sr with barite under various experimental conditions for developing a technique for the direct removal of Sr from seawater. Previous studies showed the coprecipitation of Sr by barite for the treatment of radioactive liquid waste containing Sr, but there is a lack of distribution coefficient data and the coprecipitation phenomenon is not well understood [38–40]. Coprecipitation phenomenon can be explained by two processes taking place simultaneously: (i) adsorption on the surface site and (ii) incorporation and immobilization in the structure during crystal growth [28,33]. In the present study, the following two factors were mainly investigated: (i) the affinity of the surface complex between barite surface and Sr^{2+} ion and (ii) the affinity of the substituent Sr^{2+} ion in the substituted site. The effects of pH, saturation state, ionic strength (IS), competitive ions, and barium/sulfate concentration ratio ($[\text{Ba}^{2+}]/[\text{SO}_4^{2-}]$) were investigated for determining the optimum condition for Sr removal by

using barite. Moreover, extended X-ray absorption fine structure (EXAFS) analysis was carried out to gain insights into the mechanisms involved in the uptake and retention of Sr by barite. The removal efficiency of Sr was also investigated in artificial and natural seawater for application of the remediation technique to seawater. The Fukushima Daiichi Nuclear Power Plant accident (2011) has led to serious ^{90}Sr contamination in seawater [41,42]. However, technique of the removal of ^{90}Sr from seawater have not been well developed in previous studies [43–50] because of the strong inhibition of Sr uptake by the high concentration of sodium and other competitive ions in seawater. Thus, in the present study, the difference in the Sr removal efficiency between freshwater and seawater systems was also investigated by evaluating the effect of seawater on the amount of Sr coprecipitated with barite, which allowed us to establish strategies to increase the efficiency of the technique for the direct removal of Sr from seawater. Finally, a Sr coprecipitation experiment was carried out in real seawater and the removal efficiency of Sr by using barite was evaluated under the optimum experimental conditions.

2. Materials and methods

2.1. Materials

All of the chemicals used in this study were purchased from FUJIFILM Wako pure chemical corporation, Japan, without further purification. All of the aqueous solutions tested in this study were prepared by dissolving the above chemicals in ultra-pure water prepared by a Milli-Q system (MQ; Millipore, conductivity 18.2 MΩ cm).

2.2. Experiment procedure

Barite was precipitated from a mixture of (i) Na_2SO_4 aqueous solution and (ii) BaCl_2 aqueous solution [51]. A Sr aqueous solution was added to BaCl_2 aqueous solution before addition of Na_2SO_4 aqueous solution. The pH, saturation index of barite (defined as $\text{SI} = \log(\text{IAP}/K_{\text{sp}})$ where IAP and K_{sp} are the ion activity product and solubility product of the barite, respectively), and $[\text{Ba}^{2+}]/[\text{SO}_4^{2-}]$ in the aqueous solution were fixed at pH 8.0, SI 4.2, and $[\text{Ba}^{2+}]/[\text{SO}_4^{2-}] = 1.0$, respectively, as the basic system. In the basic system, barite was synthesized by mixing 0.5 mL of 200 mM Na_2SO_4 aqueous solution in 49.5 mL of 2.0 mM BaCl_2 aqueous solution, which was subsequently shaken at 25 °C for 48 h. Additional experiments were conducted by changing one of the parameters of the basic system. The Sr concentration ($[\text{Sr}^{2+}]$) in the initial BaCl_2 aqueous solution was 0.010 mM, which was kept unsaturated with respect to celestite (SrSO_4) to prevent Sr precipitation as celestite in the basic system.

The barite precipitates and the aqueous phase were separated by filtration using a 0.20 μm membrane filter (mixed cellulose ester, Advantec, Tokyo, Japan). The solid phase was washed thrice with MQ water. The X-ray diffraction (XRD) patterns of the precipitates were measured using a powder X-ray diffractometer (MultiFlex, Rigaku Co., Tokyo, Japan), by which the mineral phase was identified by comparing the XRD patterns with those in the International Center for Diffraction Data file.

The initial and equilibrium Sr concentrations in liquid phases (mol/L) were analyzed by inductively coupled plasma-mass spectrometry (ICPMS; 7700 cs, Agilent, Tokyo, Japan). The Sr removal efficiency was calculated using the following equation:

$$\text{Removal efficiency (\%)} = \frac{(C_0 - C_e) \times 100}{C_0} \quad (1)$$

where C_0 and C_e are the initial and the equilibrium Sr concentrations in liquid phases, respectively. Sr uptake by barite ($[\text{Sr}]_{\text{B}}$; mol/kg) was calculated using the following equation:

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