



Removal of cesium ions from aqueous solutions using immobilized nickel hexacyanoferrate-sericite beads in the batch and continuous processes



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ABSTRACT

Powdered sericite impregnated into nickel hexacyanoferrate (NiHCF-sericite) was immobilized as a bead form by entrapment method using sodium alginate. The immobilized NiHCF-sericite beads have excellent mechanical strength and the increase of pressure drop caused channeling of flow was not observed through column processes even though long time operation. The existence of cesium ions onto the surface of the immobilized NiHCF-sericite beads was verified by the scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX). The maximum adsorption capacity for cesium ions of immobilized NiHCF-sericite beads was determined as the 13.877 mg/g at the initial pH 5.0 of aqueous solution and the equilibrium data followed well the linearized Langmuir isotherm equation which has the higher correlation coefficient (0.993). The breakthrough point was emerged around 50 (1.0 mL/min) and 80 bed volumes (0.5 mL/min) and the bed volumes can be still maintained as the 75 even though secondary reused immobilized NiHCF-sericite beads were used.

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Introduction

Radionuclides such as cesium, strontium and selenium are a major concern to human health if they enter the ocean, soil and air because of high toxicity. The recent big accident of Fukushima Daiichi Nuclear Power Plant, Japan in 2011 is a cause of the release a lot of radioactive substances [1]. Among them, cesium-137 with a long half-life of 30.17 years should be removed because of its high solubility and migration with aqueous media in the environment [2]. The release of cesium-137 could lead to contaminated subsurface water and/or groundwater, finally pollute the drinking water production chain, therefore, it may cause the thyroid cancer to human [3,4].

The separation/removal of cesium ions from aqueous solutions generally can be performed by means of physical-chemical methods such as coagulation-precipitation, solvent extraction, adsorption, ion exchange and membrane processes [5–7]. Among them, adsorption is a highly effective and economical process if the proper adsorbents are applied to the system that meets adsorption.

Several kinds of natural adsorbents, especially clay such as mordenite, Turkish clays and sericite have been applied to remove cesium from waters [8–11]. To increase adsorption capacity of cesium ions from waters, modified clays and synthetic adsorbents are extensively developed [12–14]. We also prepared the chemically modified sericite incorporated into nickel hexacyanoferrate (NiHCF) and successfully applied to remove cesium from aqueous solutions [15]. The incorporation of NiHCF with sericite also overcomes the difficulty of separation of NiHCF nano-particles from solutions [16]. However, powdered adsorbent is not strong enough to be applied to the continuous processes using packed-bed column operation which is widely used to the actual adsorption processes and is not acceptable pressure drop to flow [17].

Therefore, it appears in order for powdered adsorbents to be effectively used, they need to be immobilized in a bead form that conserves the adsorption capacity well and gives physical characteristics similar to those of ion exchange resins. Many immobilization methods have been suggested for applications to adsorbents and most adopted methods was entrapment and materials for entrapment were alginate, polyacrylamide, polyurethane, polyvinyl alcohol, agar and k-carrageenan. Entrapment can be achieved by mixing an adsorbent with a polyionic polymer

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material and then crosslinking the polymer with multivalent cations in an ion exchange reaction to form a lattice structure that traps the adsorbent. Especially, it is well known that biopolymer alginate has a good gel forming property, so it leads to their extensive application in biomedicine and biotechnology to immobilize or encapsulate enzymes and living cells [18,19].

In this study, powdered nickel hexacyanoferrate-sericite was immobilized as a bead form by means of entrapment method using sodium alginate. The immobilized nickel hexacyanoferrate-sericite beads were applied to the adsorption of cesium in the batch and sequential processes using packed-bed column. The results obtained from this study would be helpful for the understanding of the adsorption process and will provide possibility for application of actual treatment system which is operated by continuous processes.

Materials and methods

Materials and chemicals

Sericite used in this study was obtained from the SG mining industry located in the city of Gangneung in Korea. The preparation methods and chemical composition for chemically modified sericite with nickel hexacyanoferrate were already described in our previous work [15]. All of the chemicals were of analytical grade (Sigma–Aldrich, USA) and deionized water obtained from a Milli-Q system (Millipore, Bedford, MA, USA) was used to prepare all the solutions.

Preparation of immobilized nickel hexacyanoferrate-sericite (NiHCF-sericite) beads

The procedure to immobilize powdered nickel hexacyanoferrate-sericite (NiHCF-sericite) was as follows; some amount of NiHCF-sericite and 1 g of sodium alginate were mixed in 50 mL of deionized water at 60 °C for 2 h. The mixture was dropped into a feebly stirred 4% of CaCl₂ solution by means of syringe pump (78–8110 INT, K_d Scientific) to get uniform gel beads. The beads were aged in fresh CaCl₂ solution for 24 h and then washed with deionized water several times. Finally, the bead was dried and stored in vacuum oven at room temperature. The beads were used to remove cesium ions from aqueous solution using batch and continuous adsorption processes.

Characterization of the NiHCF-sericite beads

The physical properties for immobilized NiHCF-sericite beads were investigated. Surface area of the beads was measured by BET analysis using nitrogen gas as an adsorbate and the Khoo's method was used to measure mechanical strength [20]. Swelling ratio was also calculated by following Eq. (1):

$$\text{Swelling ratio} = \frac{(W_s - W_d)}{W_d}, \quad (1)$$

where W_s and W_d are the weight of the swollen and dry beads, respectively. Three samples were examined, respectively and average value was presented. Scanning electron microscopy (SEM, Hitachi model S-4100, Japan) and Energy dispersive X-ray spectroscopy (EDX, Oxford INCA) were employed to check surface condition and components for the before and after adsorption of cesium ions, respectively.

Batch sorption experiments

All batch sorption experiments were performed with 100 mL conical flasks sealed with rubber stoppers and placed in a rotary

shaking incubator (JEIO TECH, SI-600R, Korea) at desired temperature and time. Cesium solutions were prepared by the cesium chloride (CsCl, FW: 168.36) and the concentration was varied from 2 to 20 mg/L. The cesium sorption experiments for NiHCF-sericite contents into beads was carried out by various 0.1 g of dried beads including different mixing ratio between sodium alginate and NiHCF-sericite in 100 mL of aqueous solutions. The isothermal adsorption was performed at the desired initial cesium concentration, temperature and pH of solution. The desired pH was controlled by dilute NaOH and HCl solution and adsorption time was fixed to 24 h which is enough time to get an equilibrium state. After 24 h, the solution was centrifuged at 4000 rpm for 30 min to remove suspending NiHCF-sericite beads by centrifuge (Gyrozen, Gyro 1236 MG, Korea), and then concentration of cesium ions in supernatant was analyzed by Atomic Absorption Spectroscopy (AAS, Perkin-Elmer A Analyst 100/A Analyst 700, USA) and Inductively Coupled Plasma Optical Emission spectrometer (ICP-OES, Perkin-Elmer, UK). All sorption experiment was done three times and the average values are presented. The removal efficiency and adsorption capacity of the immobilized NiHCF-sericite beads for cesium ions were calculated as following Eqs. (2) and (3), respectively:

$$\text{Remove efficiency (\%)} = \frac{(C_i - C_f)}{C_i} \quad (2)$$

$$\text{Adsorption capacity (mg/g)} = \frac{(C_i \times V_i - C_f \times V_f)}{m} \quad (3)$$

where C_i and C_f are the initial and final cesium concentrations in aqueous solutions (mg/L), V_i and V_f are the initial and final solution volumes and m is the dry weight of the immobilized NiHCF-sericite beads (g).

Fixed-bed column sorption experiments

Continuous adsorption experiments were carried out by packed-bed column which made of borosilicate glass column (diameter = 1.0 cm, Luer-Lock made by Sigma Chemical). Column was filled with 2.0 g of NiHCF-sericite beads and cesium solutions were steadily flowed in column by up-flow direction, as is the case in most column operations. The influent cesium concentration was 20 mg/L and influent pH of aqueous solutions was fixed as 5.0 by means of diluted HNO₃ and NH₄OH. Flow rate was controlled as 0.5 (retention time: 2.4 min) and 1.0 mL/min (retention time: 1.2 min) using Acuflow Series II high-pressure liquid chromatograph (USA). Effluent samples were automatically collected by Spectra/Chrom CF-1 fraction collectors and the influent and effluent cesium concentrations were analyzed by the AAS or ICP.

Results and discussion

The beads synthesized under various mixing ratio

It is well known that adsorption capacity for metals of immobilized adsorbent was lower than that of powdered adsorbent because functional groups such as carboxyl, hydroxyl and amine groups were consumed in the immobilization processes [21]. Therefore, the amount of adsorbent itself which plays important role to bind cesium ions in beads should be increased to get high adsorption capacity for cesium ions of immobilized NiHCF-sericite beads. It was found that it could be uniformly immobilized as a bead form until the mixing ratio of 3:7 (sodium alginate:NiHCF-sericite), as summarized in Table 1. The shape and diameter of all the beads synthesized under various conditions was almost same. However, in the case of 1:9 (sodium alginate:NiHCF-sericite), the shape of the beads was not uniformly made due to the

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