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Application of recycled iron oxide for adsorptive removal of strontium

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ABSTRACT

To remove strontium (Sr) efficiently from aqueous solutions is important as Sr-90 is a radionuclide and could be a serious threat to the environment. This study examines the feasibility of Sr-90 removal using BT9, generated from a FBR-Fenton facility that was utilized in treating tannery wastewater. A rapid increased of Sr adsorption was detected when the solution pH increased from 7.11 to 11.22. A maxima adsorption capacity was estimated to be 29.85 mg Sr/g BT9 at 323 K and pH 11.22. Additionally, Sr adsorption decreases with the increase of the M/Sr from 0 to 100, indicating that the outer-sphere mechanism was importantly involved in the Sr adsorption. This Sr adsorption is spontaneous and endothermic, supported by the negative changed standard free energy with temperatures ($\Delta G^\circ = -5.75, -6.26, -6.72$ kJ/mol at 288, 303, and 323 K, respectively) and the positive ΔH° values (2.14 kJ/mol). The positive ΔS° (27.49 J/mol K) further indicates that the randomness increased at solid-solution interface during Sr adsorption.

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1. Introduction

Liquid radioactive waste (LRW) is generated during nuclear fuel cycle operations as well as the decommissioning activities in nuclear power facilities. One of the main long-lived radionuclides in the presence of LRW is the ⁹⁰Sr (28.8 years of half-life), which is produced by nuclear fission and one of the radionuclides most frequently released from LRW to the environment. Hence, removal of ⁹⁰Sr from liquids ensures health security and environmental protection. Conventionally, the LRW is treated using a series process, including precipitation, evaporation and ion exchange [1]. The treatment process can achieve satisfactory decontamination factors, but the operation costs are expensive.

Adsorption is one of the efficient techniques in the removal of radionuclides from the LRW. Inorganic materials such as, metal oxides, zeolites [2], goethite, pyrite fines, hydroxides [3,4], and clays [5] have been investigated to remove radioactive cesium and strontium ions by many researchers. The most of synthetic iron oxides were produced in powder form with nano- to micrometer particles. However, powder materials prevent the use of most synthetic adsorbents in flow process particularly due to the development of high pressure drop, as well as critical accessibility and diffusion limitations [6]. Several methods were utilized for the manufacture of adsorbents in

monolithic, immobilized, or granular forms for operation in flow systems [4,7]. Unfortunately, the transformation of powder to granular product usually has a negative influence on its performance due to the loss of active sites.

In the previous work, a recycled, granular iron oxide which was a byproduct of fluidized-bed Fenton reactor, has been demonstrated efficient for the adsorption of Cu and Pb²⁺ from wastewater [8,9]. Notably, the iron oxide (named BT1) has been further indicated to be an efficiently adsorbent for the removal of strontium ion [10]. It is well known that most of inorganic materials have selective adsorption capability for different cations. Although the BT1 material can adsorb strontium ion from water, it does not interact with cesium ion. Additionally, there are some other cations such as Mg²⁺, Ca²⁺ and Ba²⁺ may coexist with Sr²⁺ in waters (i.e. surface water, ground water and seawater). Therefore, the competitive adsorption effect between other cations and Sr²⁺ using BT material should be investigated. In this study, the iron oxide (named BT9) with higher specific surface area was employed to investigate the adsorption selectivity of strontium ions in the presence of competitive ions (i.e. Mg²⁺, Ca²⁺ and Ba²⁺) and to identify the adsorption kinetics and isotherm.

2. Materials and methods

2.1. Chemicals

All chemicals were of analytical grade and used without purification. The Sr²⁺ stock solutions were prepared using strontium chloride

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($\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$), which were purchased from J.T. Baker, USA. Nitric acid (97%) and sodium hydroxide, used to adjust the pH, were obtained from Sigma–Aldrich. All solution samples used in the adsorption experiments were prepared from de-ionized water.

2.2. Characterization of the BT9

The physical/chemical characteristics of the BT9 adsorbent used in this study were elucidated using various technologies. The crystal phases were determined by XRD (RX III, Rigaku, Japan) using graphite monochromatic copper radiation over the 2θ range 20–80°. The Brunauer–Emmett–Teller (BET) surface areas and the porosity of the adsorbents were obtained from nitrogen adsorption isotherms at 77 K using an ASAP 2010 analyzer (Micromeritics, USA). The surface morphology was examined by scanning electron microscopy (SEM, JOEL JXA-840, Hitachi S4100, Japan). Besides, the energy dispersive spectrometer (EDS) was employed to understand the distribution of the surface elements.

2.3. Batch adsorption experiments

The batch experiments were carried out in order to evaluate the adsorption processes and the equilibrium states of Sr mobilization. The Sr solution was prepared by dilution of the stock standard solution (1000 mg/L). The adsorbent, BT9, was washed ultra-pure water for several times and dried at 323 K for 24 h in an oven before further tests. All batch adsorption experiments were performed according to the following procedures: 10 mL Sr solution and a fixed amount of BT9 were poured into a 15 mL centrifuge tube. The centrifuge tubes were then put on a shaft of a rotary shaker after the caps were tightened.

To evaluate the influence of the solution pH and the competitive influence of some positive ions on the Sr adsorption, various experiments were performed by adding 0.05 g of the adsorbent into a 15 mL centrifuge tube, containing 10 mL 10 mg/L Sr solution at 303 ± 1 K. The pH of the solutions was adjusted using 0.1 N HNO_3 or NaOH solution to the designated values (7.11 ± 0.05 , 7.62 ± 0.05 , 8.01 ± 0.05 , 8.55 ± 0.05 , 9.02 ± 0.05 , 10.27 ± 0.05 , and 11.22 ± 0.05) and the competitive cations (Mg, Ca, Ba) of the solutions was designed at the ratio of M/Sr = 0, 1, 10, and 100. (M represents one of the competitive cations) All equilibrium adsorption experiments were individually conducted by shaking 0.05 g of the BT9 with 10 mg/L Sr solution using a thermostated shaker at a speed of 30 rpm for 2 h. Adsorption kinetics for Sr were measured by taking 10 mg/L Sr solution with 0.05 g of the BT9 and shaking the mixture at pH 11.22 (temperature was controlled at 288 ± 1 to 323 ± 1 K). The Sr uptake rate of q_t (mg/g) was determined by Eq. (1):

$$q_t = \frac{(C_0 - C_t) \times V}{m} \quad (1)$$

where C_0 and C_t are the metal concentration in liquid phase at the initial and at time t (mg/L), respectively; m is the adsorbent amount (g); V is the volume used in the adsorption process (L).

The Sr concentrations in the filtrate were determined by ICP-OES (iCAP 6500, Germany) at the isotope geochemical laboratory, National Cheng Kung University. The amount of Sr adsorbed on the BT9 was determined using the differences between the initial and the equilibrium concentrations in solutions. Prior to analysis, the supernatant was acidified with concentrated HNO_3 and stored in acid-washed bottles.

2.4. Adsorption isotherm

Isotherm modeling is very important for establishing an adsorption system and provides information on the capacity of the adsorbent or the amount required for removing a unit mass of pollutant under the designated conditions. The Freundlich [11] and Langmuir

isotherms [12] are the most commonly used adsorption isotherms for describing a non-linear equilibrium of adsorbate between solution and adsorbent at a fixed temperature. The batch experiments data were fitted to the isotherm models of the Langmuir and Freundlich using the least squares method. The Langmuir isotherm model is gained by combination of the adsorption and the desorption rate equations, which can be written as follows [13],

$$\frac{d\theta_t}{dt} = k_{\text{ads}}C_tN(1 - \theta_t) - k_{\text{d}}N\theta_t \quad (2)$$

where N is the maximum number of adsorption sites occupied by the Sr and θ_t is the dimensionless surface coverage ratio. When the adsorption process reaches equilibrium, Eq. (2) yields

$$q_e = \frac{K_L q_m C_e}{1 + K_L C_e} \quad (3)$$

where q_m is the maximum adsorption capacity (mg/g); q_e is the amount of Sr adsorbed at equilibrium (mg/g); and K_L is the Langmuir constant. Eq. (4) displays the rearrangement form yielded from Eq. (3).

$$\frac{C_e}{q_e} = \frac{1}{K_L q_m} + \frac{C_e}{q_m} \quad (4)$$

The Freundlich isotherm model indicates the relationship between the amount of Sr adsorbed by the BT9 (q_e , mg/g) and the equilibrium concentration of Sr (C_e , mg/L) in solution:

$$q_e = K_F C_e^{1/n} \quad (5)$$

where K_F and n are Freundlich constants that was related to the adsorption capacity and the adsorption intensity, respectively.

3. Results and discussion

3.1. Characterization of BT9

The iron oxide denoted by BT9 is a by-product, generated from a fluidized-bed Fenton facility which was utilized in treating tannery wastewater. Table 1 displays the physical/chemical properties of the adsorbent. Briefly, the XRD spectrum shown in Fig. 1 indicates the

Table 1
Physical/chemical properties of the BT9 used in this study.

Parameters	Value
Crystalline pattern	Goethite (α -FeOOH)
Particle size (mm)	0.13–0.50
Specific surface area (m^2/g)	232.31

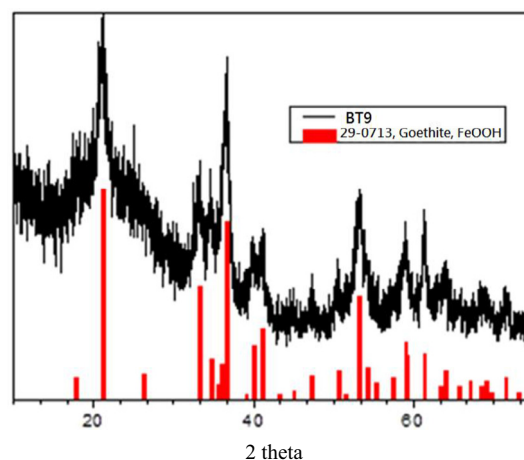


Fig. 1. XRD spectra of BT9.

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