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Adsorption and energetic heterogeneity properties of cesium ions on ion exchange resin



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ABSTRACT

The adsorption and energetic heterogeneity properties of cesium ion on ion exchange resin were investigated. The adsorption equilibrium isotherm of cesium ion on ion exchange resin could be represented by the Lamgmuir equation. Thermodynamic parameters indicated that adsorption of cesium ion on ion exchange resin was spontaneous and endothermic. Adsorption heterogeneity for the adsorption system of cesium ions on Amberlite IR 120 increased with increasing temperature and decreasing pH. A model based on linear driving force approximation was used for simulating the adsorption behavior of cesium ions in a fixed bed adsorber.

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Introduction

Radioactive waste is an inevitable residue from the use of radioactive materials in industry and the medical sector, as well as from research and nuclear establishments [1]. The management and disposal of such waste is, therefore, an issue relevant to almost all countries. Also, the development of nuclear science and technology, in particular the wide application of nuclear power, seriously threatens the human environment through radioactivity contamination. Two of the most important fission radionuclides from the reactor, 137Cs and 134Cs, are considered potentially dangerous to human health and to the environment, because the relatively high yield in nuclear power plants, long half lives and high solubility of cesium can cause its migration through ground water to the biosphere [2]. Besides, they can be easily incorporated in terrestrial and aquatic organisms, because of their chemical similarity to potassium. The bioavailability of cesium in natural systems depends on its sorption properties on solid phases [3]. The

E-mail addresses: tykim001@chonnam.ac.kr (T.Y. Kim), jinhkim@jnu.ac.kr (J.H. Kim). formation of organic and inorganic complexes does not have a significant effect on cesium speciation, and the predominant aqueous species in groundwater is the free cesium ion [4]. Different techniques, such as solvent extraction, evaporation and ion exchange, are usually used for the treatment of aqueous waste solutions containing cesium ion [5–7].

The ion exchange technique has become one of the most commonly used treatment methods for such aqueous streams, due to its simplicity, selectivity and efficiency. It is applied in waste currents, when it has a high affinity for the ion to be remove, and low affinity for other ions present in the solution. This process has been widely studied for the removal of radioactive elements from alkaline wastes, and sometimes from acid radioactive wastes [8,9].

The objective of this study is, through a series of experiments, to assess the utility of this prepared material for the removal of cesium ion from aqueous solutions, under batch and in a fixed bed mode. The relevant data, with respect to the kinetic and equilibrium of cesium ion, have been obtained using kinetic and adsorption equilibrium isotherm models. Breakthrough studies were also carried out to evaluate the effect of process parameters, such as adsorbent bed height and input concentration, on the shape of breakthrough curves.

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Theoretical models

Adsorption equilibrium

Adsorption mechanisms are so complicated, that no simple theory can adequately explain adsorption characteristics. Many expressions have been reported that describe the equilibrium relationship between the adsorbate and the adsorbent. The isotherm models of Langmuir, Freundlich and Sips were fitted, to describe the equilibrium adsorption. These equations of isotherms are given below:

Langmuir isotherm,
$$q = \frac{q_m k_L C_e}{1 + k_L C_e}$$
 (1)

where C_e is the supernatant concentration at the equilibrium state of the system (mol/m³), k_L is the Langmuir affinity constant (m³/ mol), and q_m is the maximum adsorption capacity of the material (mol/kg), assuming a monolayer of adsorbate uptaken by the adsorbent.

Freundlich isotherm,
$$q = k_F C_e^{1/n}$$
 (2)

where k_F is the Freundlich constant related to the adsorption capacity (mol/kg)(mol/m³)^{-1/n}, and *n* is the Freundlich exponent (dimensionless).

Sips isotherm,
$$q = \frac{q_m k_s C_e^{1/n}}{1 + k_s C_e^{1/n}}$$
 (3)

where k_s is the Sips constant related to the affinity constant (mol/m³)^{-1/n}, and q_m is the Sips maximum adsorption capacity (mol/kg).

Fixed bed adsorption

In order to study the adsorption behavior of cesium ions in a fixed-bed adsorber, a dynamic model was developed. The adsorption system considered is an isothermal column packed with Amberlite IR 120 at a steady state. This model includes the nonlinear adsorption isotherm, the mass balance in the liquid and solid phases, and the mass transfer resistance through the adsorbent. The model adopted here utilizes the Langmuir isotherm equation and a linear driving force (LDF) rate model, to simplify the diffusional mass transfer inside adsorbent particles. The LDF model is a lumped-parameter model for particle adsorption. A simple model for an isothermal adsorption in a fixed bed is as follows:

Mass balance:

$$-D_L \frac{\partial^2 C_i}{\partial Z^2} + v \frac{\partial C_i}{\partial Z} + \frac{\partial C_i}{\partial t} + \frac{1 - \epsilon_b}{\epsilon_b} \rho_p \frac{\partial q_i}{\partial t} = 0$$
(4)

where D_L is the axial dispersion coefficient (m²/s), v is the interstitial velocity (m/s), ε_b is the bed voidage, and ρ_p is the particle density (kg/m³).

The boundary conditions are:

$$D_L \frac{\partial C_i}{\partial Z}\Big|_{z=0} = -V(C_i|_{z=0^-} - C_i|_{z=0^+})$$
(5)

$$\frac{\partial C_i}{\partial Z}\Big|_{z=L} = 0 \tag{6}$$

where L and Z are the column length (m) and dimensionless bed height, respectively.

The associated initial conditions are:

$$C_i(Z,0) = C_0; \quad q_t(Z,0) = 0$$
 (7)

The mass-transfer rate inside particles can be represented by

$$\frac{\partial q_i}{\partial t} = k_s (q_t^* - q_t) \tag{8}$$

where k_s is the effective mass transfer coefficient (m/s), and q_t^* is the equilibrium adsorbed phase concentration.

Materials and method

Materials

A strong cation-exchange resin, Amberlite IR 120 (Rohm & Haas Co.) in hydrogen form was used for the removal of cesium ions from aqueous solution. Prior to use, the resin was washed in a column system with deionized water, to remove eventual chemicals residues. The mean particle diameter, surface area and porosity of Amberlite IR 120 were 0.75 mm, 40 m²/g and 0.62%, respectively.

Chemicals

Analytical reagent grade CsCl₂ chemical from Sigma, USA was used. Freshly prepared solutions were used throughout the experiments. Water was deionized, and purified further with a Milli-Q water purification system (Millipore, USA).

Experimental

The cesium was dissolved in deionized water to the required concentration. For adsorption equilibrium experiments, the Amberite IR 120 (0.001–0.5 g) and the cesium solution (200 cm³) were placed in a 300 cm³ flask, and then shaken for 2 days in a shaking incubator. The cesium ion concentration of the solutions was analyzed using ICP-mass (Algilent 7500CE). The amount of adsorption at equilibrium, q_e (mol/kg), was obtained as follows:

$$q_e = \frac{(C_i - C_e)V}{W} \tag{9}$$

here *q* is the equilibrium amount adsorbed on the adsorbent (mol/kg), C_i is the initial concentration of bulk fluid (mol/m³), C_e is the equilibrium concentration of the solution (mol/m³), *V* is the volume of solution (m³), and *W* is the weight of adsorbent (kg).

Adsorption column experiments were carried out in a fixed bed, which was made of a glass column of 1.2 cm in diameter and 20 cm in length. A stainless sieve was attached to the bottom of the column, followed by a layer of glass wool. A known quantity of Amberite IR 120 was packed in the column, to obtain the desired bed height of adsorbent of 7 cm. The column was then filled up with 1 mm size glass beads, in order to provide a uniform flow of the solution through the column. Cesium ion solution of known concentration, 0.75 mol/m³, was pumped upward through the column at the desired flow rate of 5.09×10^{-3} , controlled by a peristaltic pump. The cesium solutions at the outlet of the column were collected at regular time intervals for analysis, and the concentrations were determined, using ICP-mass. The column was lined with a water jacket, and the experiments were performed at 298 K.

Results and discussion

Adsorption isotherm

Adsorption equilibrium information is most important in understanding the adsorption processes. No matter how many Download English Version:

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