



Full paper/Mémoire

Evaluation of a new magnetic zeolite composite for removal of Cs⁺ and Sr²⁺ from aqueous solutions: Kinetic, equilibrium and thermodynamic studies

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ABSTRACT

In this research, a new magnetic zeolite composite (MZC) was prepared by the chemical co-precipitation of Fe²⁺ and Fe³⁺ in the presence of zeolite A. The materials were characterized by XRD, XRF, FT-IR, DTG, SEM and VSM. The capability of the composite for the removal of Cs⁺ and Sr²⁺ from aqueous solution based on magnetically assisted separation was evaluated. Adsorption studies were performed to assess the effect of relevant parameters, including pH, initial ion concentration, contact time and temperature. The kinetic data of the system were well fitted to a pseudo-second-order model, which indicates a faster kinetic sorption, by the composite. By comparison of the adsorption capacity of MZC to zeolite A, it was concluded that the iron oxide contributed to the uptake of Cs⁺ and Sr²⁺. After each adsorption experiment, the magnetic composite was efficiently separated from the solution by an easy, fast and simple magnetic separation process by a permanent magnet.

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1. Introduction

The radioactive wastes, emerging from the power nuclear reactors and the application of radionuclides in medicine, agriculture, industry and researches, have adverse effect on the human health and the environment. High fission yield and long half-life of caesium and strontium make them as the two most significant radionuclides of fission products, therefore, their separation from the radioactive wastes is important. Ion exchange; precipitation, liquid-liquid extraction and adsorption technique are the traditional methods used for the removal of caesium and strontium from liquid wastes in which the ion exchange is the most effective method [1]. Zeolites, as inorganic ion exchanger, pose high ion exchange capacity,

thermal, mechanical and radiation stability and are considered as desirable candidates for the treatment of nuclear wastes. Zeolites are crystalline aluminosilicates with three-dimensional framework structures. AlO₄ and SiO₄, tetrahedral, are linked together by sharing an oxygen atom. By the substitution of Si⁴⁺ by Al³⁺ in the structure of zeolites, a negative charge appears which is neutralized by the exchangeable cations, such as Na⁺, K⁺ and Ca²⁺. These cations are fairly mobile and could be replaced by other cations. The exchangeable cations of zeolite A are located in the alpha cage of the framework with an aperture of 4–5 Å, favorable to ion exchange [2]. After ion exchange, the adsorbent must be separated from the medium. The magnetic modification offers the advantage of efficient, easy and fast ion exchange separation by applying a magnetic field. By using micro or nano magnetic composites, a new methodology that is simpler, faster and more precise is opened for the removal and preconcentration of elements [3].

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Salah El-Din et al. loaded a zeolite with nano iron oxide [4]. Magnetite composite of clinoptilolite and urethane for the removal of Pb^{2+} was reported by Nah et al. [5]. Oliveira et al. prepared a magnetic zeolite Y composite for the removal of Cr^{3+} , Cu^{2+} and Zn^{2+} [6]. Bourlinos et al. decorated the external surface of zeolite Y with maghemite and used it for the adsorption of Hg^{2+} [7]. Several articles, concerning the application of zeolite A for metal ions and radionuclides removal, have been published [8], but no study has been reported on the synthesis and application of magnetic zeolite A composite.

In this research, we focused our attention to the synthesis of MZC to obtain the required experimental data for the adsorption of Cs^+ and Sr^{2+} . First, zeolite A was synthesized, and then MZC was prepared in ammonia solution at ambient temperature. The preparation method was simple, fast and did not need a binder polymer. The physicochemical properties of zeolite A and of the magnetic composite were characterized with XRD, XRF, FT-IR, VSM and SEM. The efficiency of the synthesized zeolite and the magnetic composite for the removal of Cs^+ and Sr^{2+} from aqueous solutions was evaluated. The effect of pH, contact time, temperature and initial concentration on the ion exchange process was investigated. The kinetic, thermodynamic and isotherm parameters of the process were also evaluated.

2. Experimental

2.1. Materials

All the chemical reagents used in this study were of analytical reagent grade [AR Grade]. $FeCl_3 \cdot 6H_2O$ (Aldrich 98%), $FeCl_2 \cdot 4H_2O$ (Aldrich 99%) and ammonium hydroxide (Aldrich 28–30% of ammonium) were used for magnetic particle preparation. Caesium and strontium were supplied as caesium chloride and strontium chloride, from Merck Company.

2.2. Synthesis of magnetite zeolite composite

Zeolite A was synthesized according to the following procedure [9]. Sodium hydroxide and sodium aluminate were dissolved in water, then stirred and boiled. The prepared solution was added to the hot solution of sodium metasilicate under vigorous stirring. After hydrothermal crystallization, the product was washed with distilled water and dried overnight in an oven at 100 °C.

MZC was prepared through the co-precipitation of Fe^{3+} and Fe^{2+} in the presence of zeolite powder. First, 100 ml of aqueous ammonia solution (1 M), were placed in a round bottom flask and a known amount of zeolite powder was added to the ammonia solution. The mixture was homogenized by using vigorously mechanical stirring and deoxygenated by bubbling of N_2 gas for 30 min. Solutions of ferric chloride (1 M in water) and ferrous chloride (2 M in HCl 2 M) with volume ratio of 4:1 were prepared and mixed together. The Fe^{3+}/Fe^{2+} solution was added dropwise to the zeolite/ammonia mixture while the mixture was vigorously stirred under N_2 atmosphere. The prepared composite was separated with the aid of a

permanent magnet. Finally, the product was washed four times with deionized–deoxygenated water and then dried at 50 °C for 5 h. Five different composites were prepared by adding different amounts of zeolite A.

2.3. Characterization of the samples

The structure of the synthesized zeolite and the magnetic composite was examined using a Bruker, D8ADVANCE X-ray diffractometer. The chemical composition of the samples was determined by a PANalytical Magix X-ray fluorescence spectrometer. FT-IR spectra were obtained by a Shimadzu, IR Prestige-21 Model spectrophotometer. Thermogravimetric analysis of the samples was conducted on Mettler, TG-50 thermal analyzer with heating rate of 10 °C/min. A vibration–sample magnetometer (Meghnatis Daghigh Kavir Co.) was used to characterize the magnetic properties of the composite. The size distribution, shape and morphology of the samples were examined by SEM. The images were taken using Seron technology AIS2100. The cation exchange capacity (CEC) of the synthesized zeolite and the magnetic composites was determined by shaking 1.0 g of the samples with 100 mL of 1 M NH_4NO_3 solution at 25 °C for 72 h. The ammonium concentration was determined by the Kjeldahl method [10]. The theoretical cation exchange capacity (TCEC) was calculated from chemical composition of the samples as a sum of exchangeable cations present in 1.0 g of zeolite.

2.4. Metal ion uptake experiments

Adsorption behavior of zeolite A and the magnetic composites was studied by batch technique. First, 0.2 g of zeolite or composite was equilibrated with 20 mL of each ion solution in a polypropylene bottle on a thermostat rotary shaker. After equilibration, the zeolite samples were separated by centrifugation while the composite separation was performed by a permanent magnet. Ion concentration was measured by ICP-AES method. The effect of pH on the cations uptake was studied by adjusting the initial pH at 3–8, using 0.1 N HNO_3 and 0.1 N NaOH solutions. For kinetic experiments, the adsorbents were contacted with ions solution for different period of time (5–120 min) at 298 K. To study the effect of temperature on the cations removal, the experiments were conducted at four different temperatures of 298, 313, 328 and 343 K, respectively. In order to investigate the effect of iron oxide content on sorption capacity, five composites with different iron oxide contents were prepared. The adsorption isotherms were studied with 0.001–0.1 N cation solutions. Cation uptake (q), expressed as the amount of ions removed per unit mass of the adsorbent was calculated according to:

$$q = (C_i - C_f) \times (V/m) \quad (1)$$

and the distribution coefficient (K_d) was also calculated as:

$$K_d = (C_i - C_f)/C_f \times (V/m) \quad (2)$$

where C_i and C_f are initial and final concentrations (mequiv/mL) respectively, m is the amount of the adsorbent (g) and V is the volume of the solution (L).

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