



Efficient removal of europium from aqueous solutions using attapulgite-iron oxide magnetic composites



Zhanhui Lu^{a,*}, Zhiqi Hao^a, Jian Wang^b, Lei Chen^{c,*}

^a School of Mathematics and Physical Science, North China Electric Power University, Beijing 102206, PR China

^b School of Environment and Chemical Engineering, North China Electric Power University, Beijing 102206, PR China

^c School of Chemical Engineering, Shandong University of Technology, 255049, Zibo, Shandong, PR China

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ABSTRACT

The attapulgite-iron oxide magnetic composites were synthesized by coprecipitation of iron oxides on attapulgite particles, and characterized by FTIR and XRD. The composites had the saturation magnetization value of 32.7 emu/g, which made the composites separate from aqueous solution easily using the magnetic separation method in large scale. The composites could preconcentrate Eu(III) efficiently from aqueous solutions. The sorption of Eu(III) on the composites was affected by pH and ionic strength obviously. The sorption isotherms were well simulated by Langmuir model, and the maximum sorption capacity was calculated to be 4.94×10^{-4} mol/g at pH 5.0, which was much higher than other materials.

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Introduction

With the rapid development of nuclear energy, large amounts of radioactive wastes are generated and some long-lived radionuclides, especially the lanthanides and actinides, are inevitably released into the natural environment. For the large volume of wastewater containing radionuclides, it is necessary to eliminate the long-lived radionuclides efficiently [1–4]. In the last decade, the elimination of the long-lived radionuclides and rare earth elements by using different kinds of polymers and their composites have been studied extensively [5–24]. The results showed that the radionuclides and the heavy metal ions can be efficiently removed by the composites from large volumes of aqueous solutions. Among the long-lived trivalent lanthanides and actinides, Eu(III) is usually taken as an analog of the trivalent lanthanides and actinides [25]. Although some lanthanides and actinides (such as ²⁴¹Am, ²⁴³Am, ²⁴³Cm, ²³⁶Np, ²⁴⁴Pu, etc.) are very important and necessarily to be investigated because of their high radioactivity and biological toxicity, they are not allowed to be used for experimental investigation in some laboratories because

of their high radioactivity [26]. Thereby, in the past decade, the interaction between Eu(III) and different kinds of natural or manmade materials have been studied extensively [27–29]. In these studies, the effects of environmental parameters such as pH, ionic strength and humic substances on Eu(III) interaction at the solid–liquid interfaces have been investigated by using batch method, surface complexation modeling and spectroscopy techniques. Fan et al. [30] investigated the sorption of Eu(III) on attapulgite by using extended X-ray absorption fine structure (EXAFS) technique, and found that the species of Eu(III) adsorbed on attapulgite surfaces were influenced by the different addition sequences of Eu(III) and humic acid to attapulgite suspension. Sun et al. [31] investigated Eu(III) interaction with sepiolite by spectroscopic and modeling, and found that the interaction of Eu(III) with sepiolite was mainly dominated by outer-sphere surface complexation at low pH and by inner-sphere surface complexation at high pH. Wang et al. [32] studied the kinetic dissociation of Eu(III) from bentonite by chelating resin and found that the fraction Eu(III) adsorbed on the irreversible sites increased with increasing pH, and the adsorbed Eu(III) could be transferred from the “weak” site to the “strong” site with increasing aging time. Kassar et al. [33] studied the sorption of Eu(III) onto anatase and rutile, and they found that the interaction of Eu(III) with titanium oxides was dependent on the crystal structures. Tan et al. [34] studied Eu(III) sorption to anatase and rutile by XPS and TRLFS

* Corresponding authors. Tel.: +86 10 61771985; fax: +86 10 61771985.

E-mail addresses: luzhanhui901@163.com (Z. Lu),

chenlei7612@163.com (L. Chen).

techniques, and found that the sorption properties of Eu(III) on anatase and rutiles were quite different. Guo et al. [35] investigated the sorption of Eu(III) on bentonite from Jinchun county (China) and simulated the sorption data with surface complexation models. Bradbury and Baeyens [36] studied Eu(III) sorption to montmorillonite, and found that the sorption of Eu(III) was dominated by the weak and strong sites of montmorillonite. The results indicated that the interaction of Eu(III) with clay minerals was mainly dominated by outer-sphere surface complexation at low pH values, and by inner-sphere surface complexation or surface (co)precipitation at high pH values.

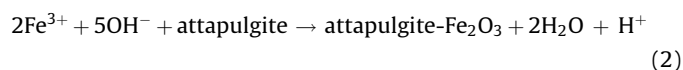
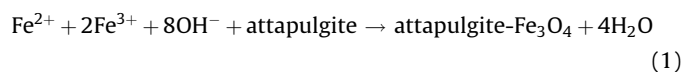
Attapulgite is a kind of hydrated magnesium aluminum silicate mineral, which consists of silica tetrahedra linked by magnesium ions in octahedral coordination, and the exchangeable cations can easily exchange heavy metal ions through ion exchange [30]. Thereby, the attapulgite can be applied to eliminate heavy metal ions from large volumes of aqueous solutions [37,38]. Although traditional methods such as filtration and centrifugation can be used to recover the attapulgite particles from aqueous solutions, it is difficult to separate the attapulgite particles from solution in large scale in real work. For example, the centrifugation method requires very high centrifugal speed and the filtration method is prone to filter blockages. Thereby, it is important to find new method to separate the attapulgite particles easily and in large scale. To solve this problem, the magnetic separation technique has been widely used because of the easy separation [39]. The solid phase can be separated from aqueous solution through magnetic separation process under the magnetic conditions, and thereby can be applied in large scale in real applications. However, based on the literature the investigation of Eu(III) sorption on attapulgite-iron oxide composites is still not available.

In this paper, we synthesized the attapulgite-iron oxide composites through coprecipitation method, and applied the attapulgite-iron oxide magnetic composites as adsorbents to remove Eu(III) ions from aqueous solutions. The composites can be separated from aqueous solutions using a simple permanent magnet easily, and thereby can be applied to remove Eu(III) ions from aqueous solutions in large scale in real applications. The interaction mechanism of Eu(III) with the attapulgite-iron oxide magnetic composites was discussed.

Experimental

Materials

The attapulgite is derived from Kaidi Co. (Gansu, China). The chemical components analyzed by X-ray fluorescence diffraction are K₂O (4.0%), CaO (4.5%), MgO (6.2%), Al₂O₃ (15.7%) and SiO₂ (47.0%) [30]. For the synthesis of attapulgite-iron oxide magnetic composites, positive ferrous and ferric ions are coprecipitated on attapulgite due to the reactions between ferrous and ferric ions and silanol groups (=Si-OH) and aluminol groups (=Al-OH) of attapulgite:



In detail, the attapulgite-iron oxide magnetic composites were synthesized as: 11.92 g FeCl₃·6H₂O and 6.12 g FeSO₄·7H₂O were added into the attapulgite suspension (2.0 g, 400 mL) at 70 °C under N₂ atmosphere. NaOH solution (20 mL, 8 mol/L) was dropwise added to form the iron oxide precipitates. Then the mixture was adjusted to pH 11 and stirred for 1 h, and aged at 70 °C

for 4 h. The sample was rinsed with Milli-Q water until the supernatant was neutral. Thus obtained composites were dried in an oven at 100 °C and used in the experiments.

Eu(III) stock solution was prepared from Eu Plasma Standard Solution for ICP-MS (Alfa Aesar). Radionuclide ¹⁵²⁺¹⁵⁴Eu(III) was used as radiotracer. The concentration of ¹⁵²⁺¹⁵⁴Eu(III) was analyzed by liquid scintillation counting using a Packard 3100 TR/AB Liquid Scintillation Analyzer (PerkinElmer). The scintillation cocktail was ULTIMA GOLD AB™ (Packard). All chemicals used in the experiments were purchased in analytic purity and used without any purification.

Sorption experiments

All the sorption experiments were carried out by using batch technique. The attapulgite-iron oxide magnetic composites, NaClO₄, and Eu(III) stock solutions were added in the test tubes to achieve the desired concentrations of different components. Before the addition of Eu(III) solution, the attapulgite-iron oxide magnetic composites and NaClO₄ were firstly equilibrated for 24 h to achieve the equilibration of Na⁺ with the composites, and then the Eu(III) solution was added into the system. The pH was adjusted by adding 0.1 or 0.01 mol/L HClO₄ or NaOH. After the suspensions were shaken for 24 h, the solid particles were separated from the solution by magnetic separation method using a permanent magnet. The amount of Eu(III) adsorbed on the solid particles was calculated from the difference between the initial concentration and the equilibrium concentration. The sorption percentage (%) was calculated as:

$$\text{sorption (\%)} = \frac{C_0 - C_{eq}}{C_0} \times 100 \quad (3)$$

where C₀ (mol/L) is the initial concentration, C_{eq} (mol/L) is the equilibration concentration.

The distribution coefficient (K_d, mL/g) was calculated using the following equation:

$$K_d = \frac{C_0 - C_{eq}}{C_{eq}} \times \frac{V}{m} \quad (4)$$

where V (mL) is the volume of the solution and m (g) is the mass of attapulgite-iron oxide magnetic composites.

Characterization

The surface functional groups of attapulgite-iron oxide magnetic composites were characterized by Fourier transformed infrared spectroscopy (FTIR) and the structures were characterized by X-ray diffraction (XRD). The FTIR spectrum was measured by a BrukerEQUINOX55 spectrometer (Nexus) in a KBr pellet at room temperature. The XRD pattern was obtained from a D/Max-rB equipped with a rotation anode using Cu Kα radiation (λ = 0.15406 nm) in the range of 5° ≤ 2θ ≤ 70°. The XRD device was operated at 40 kV and 80 mA. The magnetic properties were measured by the vibrating sample magnetometer analysis, and the magnetic curve was obtained using a model 155VSM in the range of 0 to ±12.0 kOe at room temperature.

Results and discussion

Characterization

Fig. 1 shows the XRD patterns of the bare attapulgite and the attapulgite-iron oxide composites. The characteristic peaks at 2θ values of 20.6°, 26.7° and 42.4° are found in the XRD pattern of bare attapulgite. The peaks at 2θ values of 30.1°, 35.3°,

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