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Adsorption of Europium on Al-substituted goethite

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ABSTRACT

Europium (Eu(III)) has been extensively used as an analogue of trivalent lanthanide in the radioactive wastewater from nuclear industry. The adsorption of Eu(III) on the Al-substituted goethite was investigated by characteristic and batch techniques. The characteristic results illustrated that the Al-substituted goethite was synthesized successfully and the acicular-like structure goethite was transformed to multi-domain jagged crystal with the increase of Al content, respectively. The adsorption amount of Eu(III) on Al-substituted goethite was increased with an increase of pH from 4.0 to 8.0. The thermodynamic parameters revealed that the adsorption of Eu(III) on Al-substituted goethite was a spontaneous and endothermic process. The increase of Al content increased the adsorption capacity of Al-substituted goethite to Eu(III). The maximum Eu(III) adsorption capacity was determined to be 6.75 mg/g for α-$(Fe_{0.85}Al_{0.15})OOH$ at pH = 5.5 and $T = 293$ K using Langmuir model. Furthermore, XPS (X-ray photoelectron spectroscopy) analysis verified that oxygen-containing functional groups were responsible for the major adsorption mechanism and the effect of ionic strength on the adsorption displayed the inner-sphere surface complexation of Al-substituted goethite dominated the adsorption process.

1. Introduction

With the operation of nuclear power plants, massive radionuclides, e.g. uranium U(VI), cesium Cs(I) and europium Eu(III) are produced. It may contaminate the sub-environments (soil, river, groundwater and air, etc.) and make a critical impact on human health [1,2]. Therefore, it is mandatory to remove these kinds of radioactive elements from wastewater before discharging into the environment. The adsorption technique attracted much attention due to its advantages of low operation temperature, high selectivity and high removal efficiency. In the past decades, the adsorption of Eu(III) (as a chemical analogue of trivalent lanthanides) was extensively studied with a variety of adsorbents such as natural mineral material [3–6], graphene oxide [7,8], aluminum oxide [9,10], and iron oxide [3,11,12], respectively. Among these adsorbents, natural mineral materials attracted much attention due to their characteristics of low cost and high efficiency. Hu et al. (2010) [4] reported the affinity of Na-rectorite for Eu(III) was much higher than that of Sr(II). Rabung et al. (1998) investigated the adsorption of Eu(III) on natural hematite, which elaborated the adsorption mechanism of Eu(III) on hematite. According to the above reports, natural material can be a highly potential adsorbent for the removal of radioactive nuclides in wastewater.

However, the adsorption of ions on Al-substituted goethite is rarely reported [13,14] and up to now, no report can be found for Eu(III) adsorption. Goethite, a kind of iron oxyhydroxide, is widely distributed in various compartments of the rocks and global systems. Due to its strong chemical affinity and stable structure, it can be a potentially valuable adsorbent for the sequestration of radionuclides [15–19]. Natural iron oxyhydroxides rarely exist in the form of pure phase. The iron in the crystal structure of iron oxides is universally substituted by other cations such as Al, Zn, Mn, etc. [20]. Owing to the similar atom radius and chemical valence, the substitution of Al for Fe occurred universally in natural goethite [21]. Aluminum substituted iron oxyhdroxide not only leads to a change of microstructure, but also influences the surface properties and adsorption process [22]. For example, the substitution of Al^{3+} for Fe^{3+} may lead to defect on the surface, which would have a positive impact on the adsorption capacity of Eu(III). Hence, it deserved to be investigated. Sun et al. (2014) [22] reported the sequestration of uranium on fabricated aluminum co-precipitated with goethite. However, the adsorption of Eu(III) on Al-substituted goethite was never reported.

The objectives of this study are as follows. (1) To synthesize Al-substituted goethite with different Al substitution amount and to characterize their properties using XRD, TEM. FT-IR, XPS, potentiometric titration and specific surface area; (2) to investigate the effect of pH, ionic strength, Eu(III) concentration, and adsorption time on the removal efficiencies of Eu(III) in various Al-substituted goethites; (3) to

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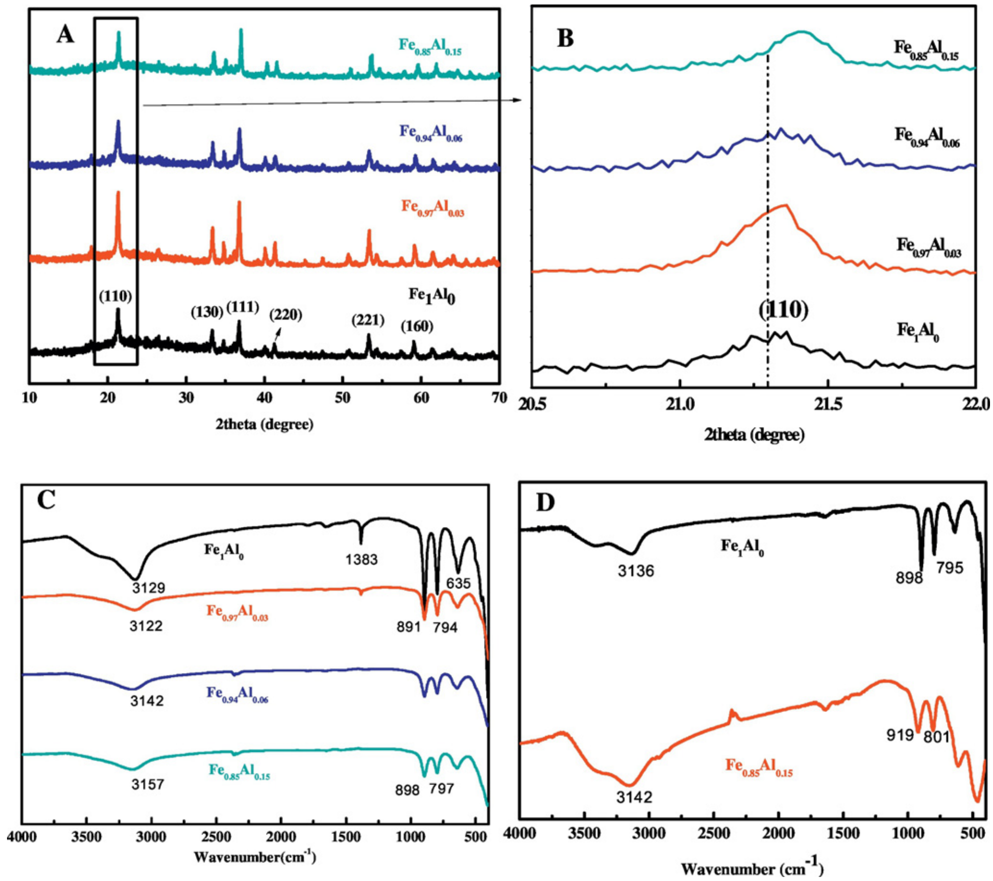

Fig. 1. Characterization of Al-substituted goethite. A: XRD patterns; B: Magnification of XRD patterns for the crystal plane of (110). C: FTIR spectra before adsorption. D: FTIR spectra after adsorption.

determine the adsorption mechanism between Al-substituted goethite and Eu(III) using XPS analysis. This study provides an innovative method for Eu(III) removal using the Al-substituted goethite in the wastewater of nuclear power plants.

2. Experimental

2.1. Synthesis of Al-substituted goethite

The Al-substituted goethite was synthesized by a rapid titration method [21]. Briefly, different mole of $Al(NO_3)_3 \cdot 9H_2O$ and

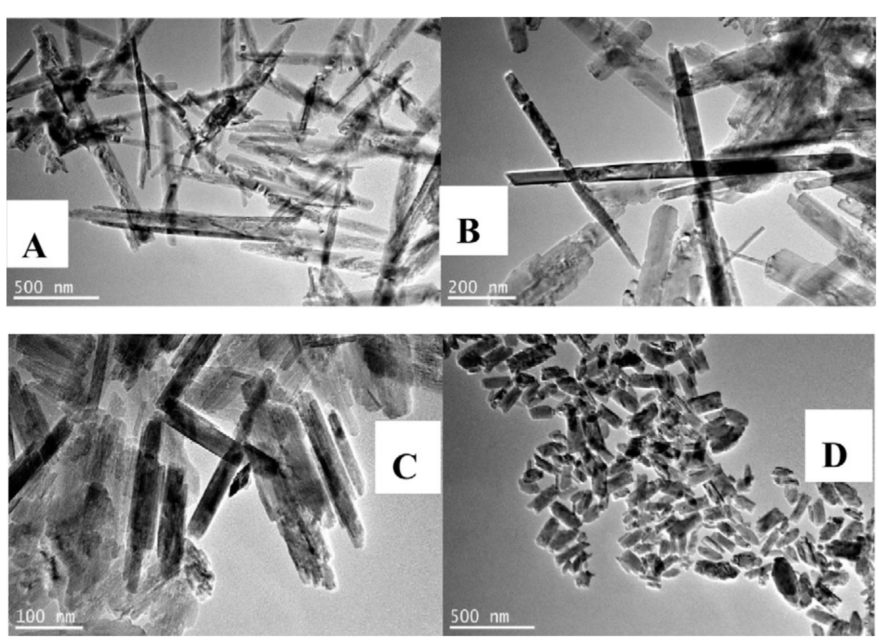

Fig. 2. The TEM images of Al-substituted goethite. A: natural goethite; B: $Fe_{0.97}Al_{0.03}$; C: $Fe_{0.94}Al_{0.06}$; D: $Fe_{0.85}Al_{0.15}$.