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Q2 Removal of Cs⁺ from water and soil by ammonium-pillared montmorillonite/Fe₃O₄ composite

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ABSTRACT

To remove cesium ions from water and soil, a novel adsorbent was synthesized by following a one-step co-precipitation method and using non-toxic raw materials. By combining ammonium-pillared montmorillonite (MMT) and magnetic nanoparticles (Fe₃O₄), an MMT/Fe₃O₄ composite was prepared and characterized. The adsorbent exhibited high selectivity of Cs⁺ and could be rapidly separated from the mixed solution under an external magnetic field. Above all, the adsorbent had high removal efficiency in cesium-contaminated samples (water and soil) and also showed good recycling performance, indicating that the MMT/Fe₃O₄ composite could be widely applied to the remediation of cesium-contaminated environments. It was observed that the pH, solid/liquid ratio and initial concentration affected adsorption capacity. In the presence of coexisting ions, the adsorption capacity decreased in the order of Ca²⁺ > Mg²⁺ > K⁺ > Na⁺, which is consistent with our theoretical prediction. The adsorption behavior of this new adsorbent could be expressed by the pseudo-second-order model and Freundlich isotherm. In addition, the adsorption mechanism of Cs⁺ was NH₄⁺ ion exchange and surface hydroxyl group coordination, with the former being more predominant.

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Introduction

In order to solve the problems of energy demand and environmental pollution, many countries are seeking new clean energy alternatives to fossil fuels. Nuclear power, which has provided approximately 18.93% of the world's electricity (International Atomic Energy Agency, 2015), is one of the most promising methods. However, nuclear power plants emit approximately 200 to 300 tons of radioactive waste globally each year (S. Yang et al., 2014; Yang et al., 2014a, b). In addition, when nuclear power accidents occur, radioactive pollutants leak into the environment. For example, in Japan's 2011 Fukushima nuclear accident, approximately 630,000–

770,000 TB radionuclides were released into the environment, including thousands of tons of radioactive waste (S. Yang et al., 2014; Yang et al., 2014a, b; Yasunari et al., 2011), which threatened human health and the ecosystems. Meanwhile, the Chinese government decided to build 6 to 8 nuclear power plants each year starting in 2016, which drew more attention to radioactive wastewater (Zhang et al., 2015). Radioactive isotopes of cesium (¹³⁴Cs and ¹³⁷Cs) are the primary concerns among radioactive wastes, because of their abundant content and a long half-life (with 2.06 and 30 years, respectively) (Zhang et al., 2015). At present, there are many ways to decontaminate radionuclide from wastewater. Precipitation, solvent extraction, membrane dialysis, and adsorption are the

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most commonly used methods (Long et al., 2013; Wu et al., 2009; Yang et al., 2016). Among these methods, adsorption is a simple operation, economical and feasible.

Clay minerals such as bentonite, illite, kaolinite, montmorillonites, sepiolite, and zeolite which are natural and low cost adsorption materials have been widely used for the removal of cesium ions from the solution (Bayülken et al., 2011; Nakano et al., 2003; Poinssot et al., 1999). Montmorillonite (MMT) belongs to the 2:1 clay family and its layered structure consists of two silica tetrahedral sheets fused to one alumina octahedral sheet. The isomorphous substitution of Al^{3+} for Si^{4+} in the tetrahedral layer and Mg^{2+} or Fe^{2+} for Al^{3+} in the octahedral layer results in a negative surface charge on the clay, and these negative sites can adsorb cations such as Ca^{2+} , K^{+} and Na^{+} between the layers through electrostatic attraction (Wu et al., 2009; Yang et al., 2016). Compared with other clay minerals, MMT has a higher adsorption capacity of cesium because of its high cation exchange capacity (CEC), large surface area, stable chemical properties and large reserves in China (J. Wang et al., 2010; T. Wang et al., 2010; Westrich et al., 1995).

Natural MMT primarily occurs in the form of Ca-MMT or Na-MMT, which has low selective adsorption to Cs^{+} . MMT pillared with ethylamine (Karamanis and Assimakopoulos, 2007; Long et al., 2013) and aluminum (Karamanis and Assimakopoulos, 2007) has been investigated, but the performance of selective adsorption of Cs^{+} was not high. The hydration radius of NH_4^{+} is similar to that of Cs^{+} . Ca-MMT pillared by NH_4^{+} would preferentially exchange Cs^{+} under other coexisting ions. In addition, MMT is usually powder-suspended in the solution, which is difficult to be separated and reused after Cs^{+} sorption. Magnetic separation, which has been used to remove cesium ions from the solution, is convenient to separate the adsorbent from wastewater (Cao et al., 2008; Fan et al., 2011; Hashemian et al., 2015; Oliveira et al., 2004; Pan et al., 2011; Sheha, 2012; Yantasee et al., 2007). MMT/ Fe_3O_4 has been used to remove heavy metals (Cu^{2+} , Pb^{2+} , and Ni^{2+}) from aqueous solutions (Kalantari et al., 2015). However, the capture of Cs^{+} in water and soil systems with the use of ammonium-pillared MMT/ Fe_3O_4 magnetic composites, and its adsorption mechanism have not been investigated.

In this study, ammonium-pillared MMT/ Fe_3O_4 was synthesized by the *in situ* co-precipitation technique. The sorption of Cs^{+} on MMT, Fe_3O_4 and MMT/ Fe_3O_4 composite was measured, and the adsorption equilibrium, kinetics, thermodynamics and their applicability in real samples were also discussed. In addition, the adsorption mechanism was investigated by means of batch experiments, adsorption model and characterization techniques.

1. Experimental section

1.1. Materials

The chemicals $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$, $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, NaCl , KCl , MgCl_2 , and CaCl_2 of A.R. grade were obtained from Sinopharm Chemical Reagent Co., Ltd., China. Ethanol, $\text{NH}_3 \cdot \text{H}_2\text{O}$ (25 wt.%) and hydrochloric acid of A.R. grade were purchased from Beijing

Beihua Fine Chemicals Co., Ltd., China. Nonradioactive cesium chloride (CsCl) of extra-pure grade was purchased from Amresco, USA. Pure water generated by a Synergy UV ultra-pure water system (Millipore, USA) was used to prepare the solution and synthesize the novel adsorbent.

1.2. Preparation of MMT/ Fe_3O_4 composite

Fe_3O_4 was synthesized by the co-precipitation method as reported previously (Jolivet et al., 2004) with a slight modification. The synthetic process of the MMT/ Fe_3O_4 magnetic particles was similar to that of Fe_3O_4 . The composite mechanism is *in situ* aggregation (Zhao et al., 2011). First, 3.1736 g of $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ (0.016 mol) was dissolved in 300 mL of deionized water; 3.00 g of MMT was added into the mixture to ultrasonic bath for 15 min, and the mixture was stirred for 30 min. Next, 7.5684 g of $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (0.028 mol) was dissolved in the suspension by stirring the mixture solution for 30 min at 80°C under nitrogen atmosphere. Then, 40 mL of $\text{NH}_3 \cdot \text{H}_2\text{O}$ was injected into the mixture rapidly, stirred under nitrogen for another hour, and cooled at room temperature. The obtained black precipitates were magnetically separated and washed repeatedly with distilled water and ethanol. Finally, the MMT/ Fe_3O_4 magnetic fluid underwent vacuum freeze-drying for 12 hr.

1.3. Characterization

Fourier transform infrared (FT-IR) spectrum was recorded with KBr over the spectral range from 4000 to 400/cm at a resolution of 4/cm (Affinity-1, Shimadzu, Japan). TGA curves were measured using a TG-DSC instrument from 25 to 800°C under N_2 at a heating rate of 20°C/min (TGA/DSC1, Mettler-Toledo, Switzerland). Powder X-ray diffraction (XRD) patterns were recorded on a powder XRD (X Pert Pro MPD, Panalytical, Holland) with $\text{Cu K}\alpha$ radiation over the 2θ range of 5°–80° at a scanning rate of 0.033°/sec. X-ray photoelectron spectrometry (XPS) was performed on a Scanning X-ray Microprobe (ESCSLAB 250Xi, ThermoFisher, USA), and the binding energy (BE) was calibrated using the C (1s) peak at 284.6 eV. The surface micromorphology was analyzed by scanning electron microscopy (SEM) equipped with energy dispersive spectroscopy (S-4800/EX-350, Hitachi, Japan).

1.4. Adsorption experiments

The adsorption behaviors of inactive cesium (cesium-133) were determined by using a batch mode of operations. A certain amount of sorbents were ground into the powdered material and then dispersed in the aqueous cesium chloride solution. After the predetermined time of shaking at 200 r/min on a rotatory shaker, the adsorbent suspended in aqueous solution was separated with a 0.22 μm cellulose acetate filter membrane. The residual concentrations of metal ions remaining in the solution were measured using an atomic absorption spectrophotometer (ICE3500, ThermoFisher, USA), and the concentration of ammonium ion was determined by UV spectrophotometer (UV-2600, Shimadzu, USA).

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