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Sorptive removal of Ce(Ⅳ) from aqueous solution by bentonite

Fengyu Huang[a], Facheng Yi[a], Zhe Wang[a,*], Haifeng Li[b]

[a] Key Subject Laboratory of National Defense for Radioactive Waste and Environmental Security, Southwest University of Science and Technology, Mianyang 621010, PR China;
[b] Institute of Engineering Surveying，Sichuan College of Architectural Technology, Deyang, Sichuan, 618000, PR China

Abstract

Bentonite has been studied extensively because of its strong adsorption capacity. A local bentonite named GMZ bentonite, collected from Gaomiaozi County (Inner Mongolia, China), was selected as the first choice of buffer/backfill material for the high-level radioactive waste repository in China. In this research, purification of raw bentonite was done to remove quartz. X-ray Diffraction(XRD) was used for the characterization of purified bentonite .In addition, the effects of contact time, solid dosage, pH, and temperature on Ce(IV) sorption to purified bentonite were also investigated by batch technique. The sorption kinetics and isotherms of bentonite for Ce(IV) indicate that the kinetic adsorption is well described by the pseudo-second-order model and the sorption isotherm is fitted well by Langmuir model. The equilibrium batch experiment data demonstrate that bentonite is effective adsorbent for the removal of Ce(IV) from aqueous solution.

1. Introduction

With the development of nuclear industry, nuclear technique has been widely used in many fields, such as national defense, industry, agriculture and medicine[1]. A large amount of radioactive waste is produced ,which contains a variety of radionuclides. The increasing level of harmful radionuclides discharged into the environment make great harm to natural environment and human society. So remove radionuclide from radioactive wastewater has special importance for all organism benefit from water. A variety of methods such as chemical precipitation[2,3], ion exchange[4] and sorption[5] are employed for removal radionuclides from aqueous solutions. A large number of domestic and international research shows, sorption is considered one of the best techniques in terms of cost,

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* Corresponding author. Tel.:+86 137 78112696
E-mail address: wz2004@126.com

simplicity of design and operation, especially for effluents with moderate and low concentrations[6].

Bentonite has attracted great interest in nuclear waste management because of its outstanding properties. The prominent properties of chemical and mechanical stability, high specific surface area and high cation exchange capacity provide bentonite retardation for the transport of radionuclides from the repository to the environment. In this context, it is of great significance for design and security assessment of the repository to study the interaction between radionuclides and bentonite. In recent years, bentonite has been greatly studied for the removal of heavy metal ions and radionuclides [7-10]. However, the sorption of Ce(IV) and other trivalent lanthanides or actinides on bentonite is still scarce.

Ce(IV) has similar chemical properties with radioactive elements Pu(IV) because the ionic radius of Ce(IV) is almost the same for the trivalent lanthanides and actinides. The sorption of Ce(IV) on activated carbon [11], zeolite[12] had been studied, but no available study was focused on the sorption behaviors of Ce(IV) on bentonite. The main purposes of this work are: (1) prepare purified bentonite and characterize using X-ray Diffraction (XRD); (2) the effect of pH, contact time and temperature on Ce(IV) sorption on bentonite was studied in detail;(3) calculate the thermodynamic parameters of Ce(IV) adsorption on bentonite, such as ΔH°, ΔS° and ΔG°; (4) to discuss the sorption mechanism of Ce(IV) on bentonite.

2. Experimental

2.1. Materials

All reagents used in this study were of analytical grad reagents. Milli-Q water was used in all experiments. Ce(IV) stock solution was prepared by dissolving weighed amounts of ceric ammonium nitrate in Milli-Q water.

2.2. Purification of raw bentonite

The main aim of this purification is to remove impurities. This process was done using the following steps: at the beginning, 10g nature bentonite was mixed with Milli-Q water in proportion of 1:10 by weight and stirred with a high-speed (2000rpm) for 2 h. The suspensions standing for 24h, then upper suspensions were centrifuge at 7000rpm.The obtained solid was dried at 105℃for 2h in order to remove free water. At last, the sample was milled through a 200-mesh screen and used in the experiments.

2.3. Characterization

Structural characterization of bentonite was carried out by X-ray diffraction (XRD) in a diffractometer (X'Pert PRO, PANalytical, Holland) equipped with a Cu Ka radiation source between 3° and 80° at room temperature. The JCPDS PDF database was used for the phase identification.

2.4. Adsorption experiments

The sorption of Ce(IV) on bentonite was investigated by using batch technique in 250 mL stoppered conical flask under ambient conditions. The stock suspension of bentonite and $NaClO_4$ were pre-equilibrated for 24 h, then Ce(IV) stock solution was added in bentonite suspension to achieve the desired concentrations of different components. The mixtures were shaken by a constant-temperature shaker (IS-RDH1, Crystal, America) and then were centrifuged (TG1850-WS, Luxiangyi, Shanghai, China) at 7000 rpm for 30 min. The kinetics experiment were performed by mixing 0.1g bentonite with solutions of Ce(IV) (2.26 mmol/L).The mixtures were separated after shaking 0–24 h at 303 ± 1 K. Except when pH effect was studied, all experiments were carried out at initial pH = 1.8 $\pm$ 0.1. The pH was adjusted with 0.1 M perchloric acid and 0.01 M sodium hydroxide. In addition, the pH of the system was determined during the sorption process using an pH meter (S975 SevenExcellence, Mettler Toledo, Shanghai, China). Sorption isotherms were performed (T = 288, 303, 318 and333 $\pm$ 1 K), with an initial Ce(IV) concentration ranging from 1.52 to 4.28 mmol/L. The amounts of absorbed Ce(IV) were calculated from the difference between the equilibrium concentration (C_e) and the initial one (C_0), i.e., $C_s=(C_0−C_e)V/m$, where m (g) is the mass of bentonite and V (L) is the volume of the suspensions. The adsorption percentage