



Removal of uranium from aqueous solution by a low cost and high-efficient adsorbent

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ABSTRACT

In this study, a low-cost and high-efficient carbonaceous adsorbent (HTC-COOH) with carboxylic groups was developed for U(VI) removal from aqueous solution compared with the pristine hydrothermal carbon (HTC). The structure and chemical properties of resultant adsorbents were characterized by Scanning electron microscope (SEM), N₂ adsorption–desorption, Fourier transform-infrared spectra (FT-IR) and acid–base titration. The key factors (solution pH, contact time, initial U(VI) concentrations and temperature) affected the adsorption of U(VI) on adsorbents were investigated using batch experiments. The adsorption of U(VI) on HTC and HTC-COOH was pH-dependent, and increased with temperature and initial ion concentration. The adsorption equilibrium of U(VI) on adsorbents was well defined by the Langmuir isothermal equation, and the monolayer adsorption capacity of HTC-COOH was found to be 205.8 mg/g. The kinetics of adsorption was very in accordance with the pseudo-second-order rate model. The adsorption processes of U(VI) on HTC and HTC-COOH were endothermic and spontaneous in nature according to the thermodynamics of adsorption. Furthermore, HTC-COOH could selectively adsorption of U(VI) in aqueous solution containing co-existing ions (Mg²⁺, Co²⁺, Ni²⁺, Zn²⁺ and Mn²⁺). From the results of the experiments, it is found that the HTC-COOH is a potential adsorbent for effective removal of U(VI) from polluted water.

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1. Introduction

Uranium is particularly crucial for being used as the raw material of nuclear power plants. With the rapid development of nuclear energy, large doses of uranium contamination have been released into the water environment associated with nuclear industry, especially uranium ore mining and hydrometallurgy, etc. The uptake of uranium by human beings would cause serious health problems, such as severe liver damage, kidney damage and eventually death due to its extremely chemical and radioactive toxicity [1]. For the above reasons, the removal of U(VI) from aqueous solution is of great importance in consideration of the dual significances in the protection of environment and alleviation of the shortage of the nuclear material resource.

Several techniques have been developed for separation uranium from the wastewater, such as chemical precipitation [2], ion

exchange [3] and adsorption [4–6]. Lately, adsorption, due to its high-efficiency and easily handling [7] has been gradually utilized to this area. Therefore, the design and preparation of adsorbents with low cost, stability and high-efficiency and selectivity has been drawing considerable interests in processing uranium contaminated wastewater in recent decades.

It has been reported that carbonaceous materials produced by hydrothermal carbonization of biomass showed higher surface area and novel porous property [8]. Liu and Zhang [9] synthesized a hydrothermal char and it exhibited high adsorption capacity toward phenol and Cu(II) from aqueous solution, however, the temperature of hydrothermal carbonization was high up to 300 °C, and the high temperature is not appropriate to acquire the oxygen-containing functional groups on the surface. Recently, Titirici et al. [10] have firstly synthesized a mesoporous carbon by hydrothermal carbonizing crude plant material at a more mild temperature (<200 °C), and the products showed their apparent pore sizes between 10 nm and 100 nm.

And to the best of our knowledge, the adsorbents with hydrophilic surface and more acid oxygen-containing groups are

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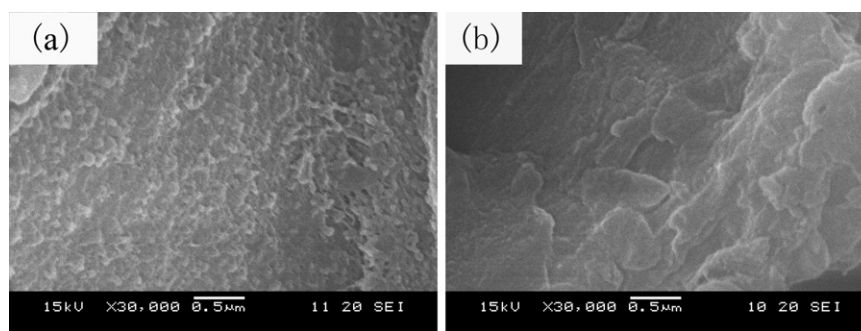


Fig. 1. The SEM of HTC(a) and HTC-COOH(b).

more suitable for adsorption of nuclide ions from aqueous solution. There are various adsorbents with carboxylic groups which is hydrophilic have applied in adsorption of U(VI) in the aqueous solution [4,11,12]. These adsorbents with carboxylic groups show good adsorption capacities toward U(VI). Therefore, in this work, the hydrothermal carbon (HTC) synthesized at 200 °C was oxidized by 2 mol/L HNO₃ solution to obtain carboxyl-rich hydrothermal carbon (HTC-COOH) to improve the sorption capacity and selectivity toward uranyl ions. The efficiency of HTC-COOH for removal of uranium from aqueous solutions was investigated compared with pristine HTC, and scanning electron microscope (SEM), N₂ adsorption–desorption, and Fourier transform-infrared spectra (FT-IR) and acid–base titration were used to characterize the structure and textural property of HTC and HTC-COOH. The effects of solution pH, contact time, initial concentrations and temperature on U(VI) adsorption onto HTC and HTC-COOH, as well as adsorption kinetics, isotherm models, and thermodynamics were studied. And the selectivity toward U(VI) was also investigated in aqueous solution containing Mg²⁺, Co²⁺, Ni²⁺, Zn²⁺ and Mn²⁺.

2. Experiment

2.1. Materials

For the preparation of U(VI) stock solution (1 g/L), an accurate amount (0.1179 g) of U₃O₈ was dissolved by hydrochloric acid ($\rho = 1.18$ g/mL) and hydrogen peroxide (30%). And the solution was evaporated to near dryness and then transferred to a 100 mL volumetric flask. A series of standard U(VI) solutions with concentrations range from 10 to 110 mg/L were prepared by successive diluting the stock solution by deionized water. All the other chemicals and agents without special note were analytical grade purchased from the Shanghai Chemical reagent corporation.

2.2. Synthesis of carboxyl-rich hydrothermal carbon

The pine needles were collected from the author's campus, and washed with deionized water for several times, dried at 80 °C and crushed into powder. 3.0 g pine needle powder was added into a 50 mL volume capacity Teflon-lined stainless steel autoclave containing 0.1 g citric acid and 30 mL of deionized water. The autoclave was sealed and heated at 200 °C for 16 h and then cooled to room temperature. The products were filtered off and washed with deionized water and ethanol for several times, and finally dried at 60 °C in a vacuum oven and designated as HTC.

In order to obtain carboxyl-rich hydrothermal carbon, 0.5 g HTC was reacted with 80 mL 2 mol/L HNO₃ aqueous solutions at 353 K for 1 h under vigorous stirring and refluxing. The products were washed thoroughly with deionized water until the pH was close to neutral, and then dried at 80 °C under vacuum and designated as HTC-COOH.

2.3. Characterization

The surface functional groups of hydrothermal carbons were characterized by FT-IR with KBr pellet on a Nicolet i-380 instrument within the range of 500–3600 cm⁻¹. Surface morphology was characterized on a JEOL JSM-5900 scanning electron microscopy. N₂ adsorption–desorption isotherms of HTC and HTC-COOH was conducted at 77 K by a Micromeritics ASAP 2020 analyzer. To determine the carboxylic groups on the surface of adsorbents we prepared, an acid–base titration is used [13]. Typically, 20 mL NaOH solution (0.0201 mol/L) was added into a Erlenmeyer flask with 0.100 g of adsorbent. The flasks were shaken for 4.0 h for sufficiently contact. And the filtrate of the mixture was titrated to the pH of 7.0 with a pH meter (PHS-3C).

2.4. Batch adsorption experiments

Adsorption behaviors of hydrothermal carbon were carried out in a temperature controlled water bath shaker. In a typical procedure, 0.01 g of adsorbent was added into a flask containing 50 mL U(VI) solution at a fixed pH (adjusted by 0.1 mol/L HCl and NaOH solution), concentration and temperature. After shaking for several hours, the mixtures were centrifuged prior to measurement of the concentrations of the final solutions by arsenazo-III spectrophotometry method [14] on a UV–vis spectrophotometer at 660 nm. The coexisted ions such as Na⁺, Mg²⁺, Co²⁺, Mn²⁺ and Ni²⁺ were obtained by an ICP-OES (ICPE-9000, Shimadzu). The amounts adsorbed (q_e , mg/g) on the sorbents were calculated according to the Eq. (1).

$$q_e = \frac{(C_0 - C_f)V}{m} \quad (1)$$

where C_0 and C_f (mg/L) are the initial and final metal ions concentrations in the aqueous solution, respectively. V the volume of the solution (L), m is the mass of dry adsorbents used (g).

3. Results and discussion

3.1. Characterization

According to the SEM micrographs in Fig. 1, the rough surface and lots of carbon particles were observed in the surface of HTC, demonstrating the pine needles were hydrolyzed and carbonized. The lower carbon particles and smoother surface in the HTC-COOH may be due to the decomposition by the HNO₃ solution. The oxidation process of carbonaceous materials with HNO₃ solution may be shown as following: $C + 4HNO_3 = CO_2 + 4NO_2 + 2H_2O$, indicating that the carbon material will be decomposed lost weight in the oxidation [15], which may lead to smooth surface of HTC-COOH. In addition, the similar phenomenon is observed by Baniamerian et al. [16] who attribute the decrease of the length of mesoporous

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