Interaction of U(VI) with amine-modified peanut shell studied by macroscopic and microscopic spectroscopy analysis

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ABSTRACT

Peanut shell, a widely available agricultural waste, was successfully grafted with quaternary ammine groups on its surface by using amination modification. The sorption mechanism of U(VI) on the amine cross-linked peanut shell (PNN) was examined by batch experiments and spectroscopy analysis. The physical adsorption was dominated by the liquid film diffusion and the intra-particle diffusion, whereas the chemical adsorption was dominated by surface complexation with quaternary amine groups and carboxyl groups. The maximum adsorption capacity of U(VI) on PNN was calculated to be 574.81 mg g^{-1} at pH = 6.0 and T = 298 K, which was much higher than that of U(VI) on pristine pure peanut shell and many other materials. Consideration of the high sorption efficiency and eco-friendliness of peanut shell, PNN presented promising potential in the preconcentration of radionuclides in radioactive pollution management.

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1. Introduction

Uranium, a representative actinide element, has attracted worldwide attention due to its high radioactivity, biotoxicity, and mobility in the aqueous system (Liu et al., 2017). With the development of nuclear power, large amounts of uranium-containing wastewater from weapon manufacturing activities and nuclear plant have been discharged into the external environment (Dong et al., 2017). Most of uranium species exist in the form of hexavalent state (U(VI)) in the aquatic environment, such as UO_2^{2+} ions, which are soluble and easy to form U(VI) complexes. The U(VI) species are difficult to be degraded in the aquatic environment, easily deposited in human body and accumulated through the food chain (Liu et al., 2018). Therefore, U(VI) species have great potential toxicity and appear carcinogenic hazardous to aquatic organism and human health. In view of this, it is crucial to develop an efficient technology to eliminate U(VI) from contaminated water (Hu et al., 2016). Multiple technologies have been employed for U(VI) removal from aquatic environment (Yao et al., 2018). Compared among these technologies, adsorption has been widely accepted as the most promising one, owing to its simple operation, superior remediation effect, and widely adaptability (Hu et al., 2017a; Zhao et al., 2017a). Hence, tremendous efforts have been made to synthesize effective sorbents with excellent sorption capacity for widespread applications in radionuclide treatment.

Recently, commercial chemosynthesis materials, such as montmorillonite (Chen et al., 2015), graphene oxides (Liu et al., 2015; Wang et al., 2016; Zhao et al., 2015), metal organic framework materials (Li et al., 2018; Yao et al., 2018), bentonite clay (Anirudhan and Ramachandran, 2006) nanotubes (Hu et al., 2017a; Linghu et al., 2017a; Sun et al., 2016a; Zhu et al., 2018) and modified carbon nitride composites (Yu et al., 2018; Zou et al., 2016) have been extensively used as adsorbents for the removal of radionuclides from aquatic environmental solutions. Excellent properties of these materials, such as high surface area, favorable porous structure, remarkable mechanical/thermal stability, and high adsorption capacity, have drawn a great deal of attention (Hu et al., 2017b).

However, common issues involved in the synthesis process, as secondary contamination, complicated synthetic procedure, and expensive precursors, have significantly hindered their practical application in environmental pollution treatment (Chen et al., 2018; Gu et al., 2018; Hu et al., 2017c). Since the biorefinery concept was proposed by Fernando et al. in the year of 2006 (Fernando et al., 2006), the secondary utilization of biomaterials have boomed and drawn a great attention. As a type of environmental benign materials, porous biomaterials contain plenty of cellulose, hemicellulose, lignocellulose, proteins, fats, and other chemicals (such as vitamins, dyes, and flavors). Cellulose structures have large amounts of oxygen-containing functional groups, which can chelate toxic cations by ion exchange or complexation (Anirudhan and Radhakrishnan, 2009; Sud et al., 2008). The superior thermal and chemical stability ensure the biomaterials to be a kind of potential adsorbent (Liu et al., 2017; Xu et al., 2011a). Up to now, a wide variety of agriculture by-products, such as coconut coir pith (Anirudhan et al., 2009), pine wood (Wang et al., 2017), tamarind fruit shell (Anirudhan and Radhakrishnan, 2009), and wheat straw (Xu et al., 2011b) have been studied to eliminate pollutants from wastewater. Peanut shell (PN) is one kind of porous biomaterials and its output in China approximately accounts for one third of the gross production all over the world. Furthermore, the PN contains more than 60% cellulose and hemicellulose, which makes it easy to be grafted with functional groups (Cheng et al., 2016). To date, most reported biomaterials usually exhibited well physiochemical properties but low adsorption capacities. Hence, it is necessary to design and fabricate a kind of efficiency bio-sorbent with large sorption capacity, fast sorption rate, and long-term stability.

In order to improve the adsorption performance of sorbent, various modification methods have been applied to functionalize the raw materials (Anirudhan et al., 2009; Cheng et al., 2016; Li et al., 2017). Among all, Schiff-base reaction, which introduces quaternary ammonium compounds $[(N^+)(CH_2CH_3)_3Cl^-]$ onto the surface of raw material, is conducted by esterification reaction and amination reaction (Shang et al., 2017a). These reaction processes could yield the imide functional groups (C=N) between the amine groups and the aldehyde groups. It has been successfully realized by the amalgamation of DMAP/HOBT, EDC/NHS, and DCC/DMAP (Pan et al., 2017). Quaternary ammonium groups have both high adsorption capacities and good selectivity toward metal ions, owning to the existence of multiple donor atoms (e.g., N and O atoms) (Chen et al., 2016; Li et al., 2016). In addition, Schiff-based adsorbents also showed strong influence on the polarity and the dispersibility of bio-adsorbent (Prolongo et al., 2018). The decoration of quaternary ammonium groups on GO were also surveyed and exhibited good adsorption performance (Pan et al., 2017; Zhao et al., 2017b). It has been demonstrated that the implantation of quaternary ammonium groups via the amination modification could effectively increase the inter plane forces of GO nanosheets, inhibited the aggregation and improved the hydrophobic character. However, no report was available on the amine cross linked PN as a bio-adsorbent for U(VI) removal from aquatic environment. In addition, the interaction mechanism of these amine cross linked PN with U(VI) still remained to be explored for the application in radioactive wastewater treatment.

Herein, we investigate U(VI) sorption on pristine PN and amine cross-linked PN (PNN) by batch technique and spectroscopy analysis. The objectives of this paper are as follows: (1) to effectively synthesize PNN and examine its morphology and microstructure by transmission (TEM), scanning electron microscopy (SEM), X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), and X-ray photoelectron spectroscopy (XPS); (2) to evaluate the sorption of U(VI) on PNN as the influence of contact time,

temperature, pH, and ionic strength by batch technique; and (3) to interpret the removal mechanism of U(VI) with PNN by XPS, FTIR, and SEM-EDS techniques. Herein, the adsorption processes of U(VI) onto PNN are researched from the macroscopic to the microscopic level.

2. Experimental

2.1. Chemicals and materials

The pristine PN, obtained from local market in Beijing, was used as the raw feedstock. The PN was firstly rinsed with tap water and thoroughly dried in an oven at 70 °C for 24 h to constant weight. After being smashed, it was sieved through a 300-mesh sieve. The material was washed with Milli-Q, dried until constant weight, and stored for further use. Analytical grade reagents were obtained from Sinopharm Company and used as received without any further purification. The preparation of U(VI) stock solution (600 mg L^{-1}) was carried out by dissolving $UO_2(NO_3)_2 \cdot 6H_2O$ with Milli-Q water.

The PN (3.0 g) was treated with epichlorohydrin (6 mL) and N,N-dimethylformamide (5.0 mL) for 60 min in a 40 mL conical flask at 85 °C. After the addition of edetic acid (EDTA) (2.0 mL), the solution was continually stirred for 45 min at 80 °C. Next, 40% trimethylamine (w/w) (5.0 mL) was added and further stirred for 120 min at 85 °C. The products were rinsed with Milli-Q water to remove the residual chemicals on the surface, then dried at 60 °C for 24 h, and sieved to obtain targeted particles with the grain diameter <50 μm. PNNs were collected and then used in the following experiments. The synthesis processes of PNN were shown in the Supporting Information.

2.2. Characterization

The PN and PNN samples were characterized by SEM (S-4800 Hitachi), TEM (JEM-1011), XRD (Dmax/RB diffractometer), FTIR (Bruker Tensor-27 spectrophotometer), N$_2$-BET (Tristar II Brunauer-emmett-teller), and XPS (Thermo Escalab 250) techniques. The XRD patterns were recorded with a reflection mode (Cu Kα source, $\lambda = 1.5412$ Å) in $2\theta = 5°-70°$ with a scanning rate of 6° min^{-1}. FTIR spectroscopy analysis was performed in pressed KBr pellets (Aldrich, 99%) over the range of 400–4000 cm^{-1} at room temperature. The XPS spectra were conducted with monochromatized Al Kα source. C 1s peak at 284.8 eV was recorded as a reference. After the background subtraction, the recorded lines were fitted by the XPSPEAK41 program.

2.3. Batch experiments

The U(VI) adsorption experiments on PN and PNN were implemented in the polyethylene tubes under ambient conditions. Prior to the addition of U(VI) solution, the PN or PNN stock suspensions (6.0 g t^{-1}) were pre-mixed with NaNO$_3$ for 24 h to achieve equilibration with NaNO$_3$. In addition, NaNO$_3$ solution with the concentration of 0.01 mol L^{-1} was used to imitate the comparable ionic strength in the actual aquatic environment. The pH was adjusted by adding negligible volumes of 0.01 or 0.10 mol L^{-1} HNO$_3$ or NaOH solutions. The U(VI) concentration was measured with the Arsenazo-III method using a UV−vis spectrophotometer (UV-2550) at the wavelength of 650 nm. The adsorption percentage ($R\% = (C_0$-$C_e)/C_0 \times 100\%$) and the adsorption capacity of U(VI) adsorbed on PNN ($q_t = (C_0$-$C_e) \times V/m$) were calculated from the mass of solid (m, g), and the volume of the suspension (V, mL), the initial U(VI) concentration (C_0, mg·L^{-1}), and the equilibrium concentration (C_e, mg·L^{-1}). The data were averaged from triplicate tests and the error