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Investigation of uranium (VI) adsorption by poly(dopamine) functionalized waste paper derived carbon

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ABSTRACT

Waste paper derived carbon coated with poly(dopamine) (WPC@PDA) as a novel adsorbent was synthesized successfully by activation of KOH and carbonization of waste A4 paper, followed by *in situ* polymerization of dopamine. WPC@PDA composites possessed a 3D hierarchically interconnected porous structure and the Brunauer–Emmett–Teller (BET) specific surface area is calculated to be 364.6 m^2/g. The enrichment of uranium for WPC@PDA composites from not only aqueous solution (mg/L) but also simulated seawater (μg/L) was fully evaluated under different scenarios. The experimental results indicated the maximum absorption capacity to uranium (VI) of WPC@PDA composites was 384.6 mg/g at pH 7. In addition, the distribution coefficient (K_d) of U (VI) ion with other competing ions for WPC@PDA composites is 1.97×10^4 mg/L and demonstrated excellent selectivity to uranium (VI) in simulated seawater, which augurs well for the application of the adsorbent in seawater.

1. Introduction

With the increasing depletion of traditional energy, the growing development of renewable energy has attracted widespread interest. Nuclear energy as a kind of clean energy is progressing rapidly. Uranium is an important nuclear fuel and the requirement for uranium is increasing. To meet this demand, the researchers put the focus on the ocean. There are about 4 billion tons of uranium in seawater, which provides a huge resource base [1]. However, a challenge exists to extract uranium from seawater with high salinity and extremely low uranium concentration (3.3 μg/L) [2].

In the past, various preconcentration methods such as flotation [3], electrochemical treatment [4], co-precipitation [5] and adsorption [6–8] have been applied for the removal of uranium. In particular, adsorption as the most promising approach emerges for extracting uranium from seawater because of its potential advantages of high efficiency, low cost and easy accessibility [9]. Important criteria for the evaluation of an adsorbent to remove uranium are adsorption capacity and selectivity. Carbon materials have been widely studied because of the more activated sites and high surface areas [10]. In addition, the materials display different carbon nanostructures such as mesoporous carbons, carbon nanotubes and carbon nanospheres. For example, Wang et al. found that the ordered mesoporous polymer-carbon composites exhibited excellent adsorption performance for U(VI) [11]. Tran et al. reported that Br-PADAP-impregnated multiwall carbon nanotubes as the adsorbent for the efficient enrichment of U(VI) [12]. Li et al. fabricated amino functionalized flake graphite composite and applied them as adsorbents for capturing U(VI) [13]. Zare et al. reported AgOH–MWCNTs nanoparticles as adsorbents to remove uranyl ions from aqueous solutions [14]. However, the relatively high cost limits these carbon materials in their application. Therefore, the preparation of carbon materials from inexpensive, abundant, and sustainable materials as highly selective uranium adsorbents is worth investigating.

Waste paper derived carbon (WPC) is a cheap and readily available raw material, which has been selected with a special focus on upgrading the waste for utilization. WPC is produced by chemical activation which, in generally, is a priority route because of its high specific surface area, abundant resources, environment-friendly and low cost [15]. In order to improve the porosity of the material, we have selected KOH as the porogen to activate the surface of the material. Three-dimensional porous carbon material

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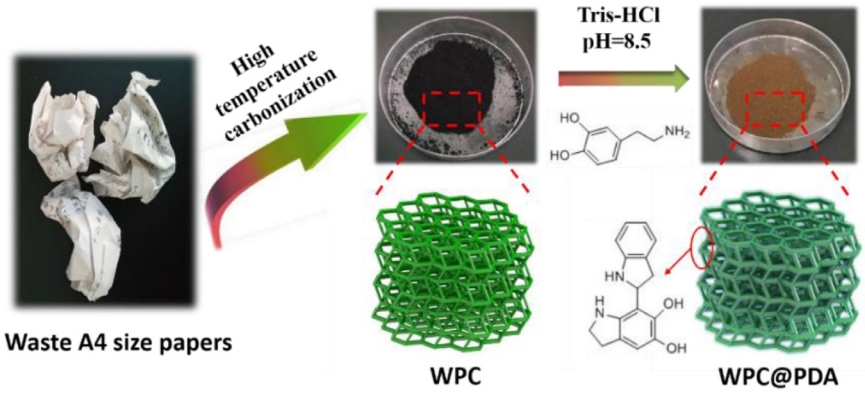

Scheme 1. Proposed the synthesis route of WPC@PDA.

with hierarchical pores is synthesized by the high temperature carbonization. In addition, in order to improve the selectivity for uranium ions, we have built the functionalized modifications on the surface of the material.

Dopamine, a remarkable biopolymer, is secreted by mussels and easily oxidized to dopaquinone, and then cross linked self-polymerization through dismutation reaction, which provided a strong attachment onto the substrate, as well as the generation of polydopamine(PDA) [16]. PDA has aroused widespread attention due to its strong adhesion, favorable biocompatibility and rich functional groups. There are abundant catechol and imine/amine groups in the PDA structure, which can well improve the substrate's hydrophilicity and provide active sites for the binding to heavy metal ions [17]. For instance, researchers have attempted to modify SBA-15 [18] and graphene oxide [19] by polydopamine. However, the adsorption ability of PDA modified adsorbent for low-concentration uranium was not demonstrated in these studies. In our study, we investigated one possible application for extracting uranium from simulated seawater (concentration of uranium ~3.3 μg/L) and provided the theoretical basis for oceanic uranyl extraction.

Herein we report on the synthesis of hierarchically porous carbon material as the precursor and the employment of PDA for surface modification by *in situ* polymerization form a polydopamine coating (WPC@PDA), which was applied to adsorb uranium (VI) from the aqueous solution simulated seawater. The WPC@PDA composites were characterized by SEM, FTIR, XRD, TEM, BET and XPS. Additionally, the influence of the solution pH, initial uranium (VI) concentration, temperature, and contact time on the adsorption of uranium (VI) were discussed in detail. Furthermore, the adsorption of uranium (VI) from simulated seawater containing coexisting ions and trace uranium (VI) were conducted. By a series of adsorption tests, WPC@PDA shows outstanding adsorption capacity and excellent selectivity for the extraction of uranium.

2. Experimental section

2.1. Synthesis of WPC@PDA

The preparation of WPC materials is placed in the support information. WPC was dispersed in Tris buffer solution (25 mM, pH = 8.5) by magnetic stirring for 20 min, and then 20 mM dopamine hydrochloride was added. The coating process was maintained at 25 °C for 24 h. The obtained PDA modified WPC composites were separated by centrifugation and washed with distilled water. Finally, the WPC@PDA composites were dried in a furnace (60 °C) for use. The detailed experimental procedure for adsorption of uranium is recorded in the support information.

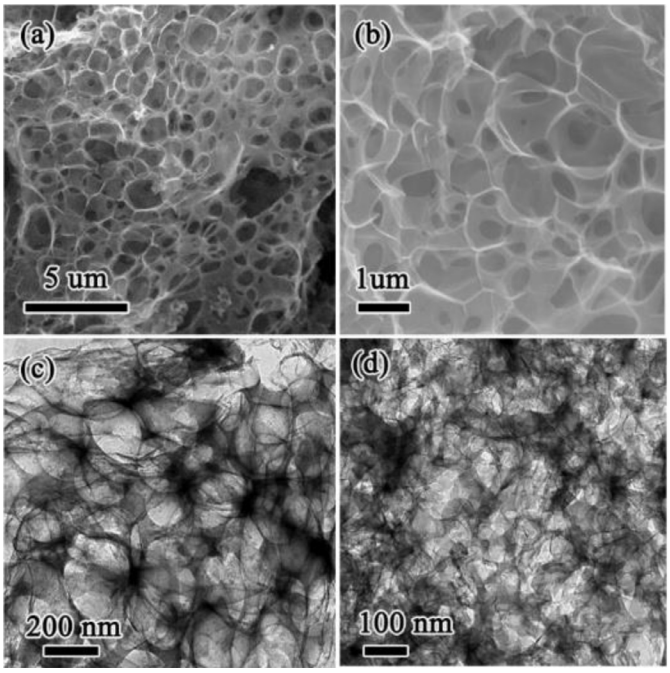

Fig. 1. SEM images of WPC (a) and WPC@PDA (b), TEM images of WPC (c) and WPC@PDA (d).

3. Results and discussion

3.1. Characterization of samples

The synthesis of WPC and WPC@PDA are described in Scheme 1. The hydrothermal carbonization helps in introducing the aromatic rings and removes other impurities present in the precursor [20]. In addition, the hydrothermal treatment also results in the materials carbonization under the high pressure condition. KOH was used as the porogen to provide more pore structure for the material, which supplied an open freeway for the diffusion of uranium ions. WPC@PDA were prepared by the self-polymerization of dopamine onto the surface of WPC.

The SEM images in Fig. 1(a) and (b) show an interconnected porous structure of WPC and WPC@PDA. It is evident that the continuous pores overlap each other, which form the interconnected network with ultrathin carbon walls. Transmission electron microscopy (TEM) images of WPC and WPC@PDA indicate that the walls of the nanopores are thin enough to exhibit bright

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