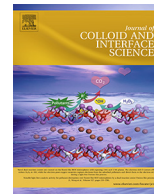




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Easy-handling bamboo-like polypyrrole nanofibrous mats with high adsorption capacity for hexavalent chromium removal



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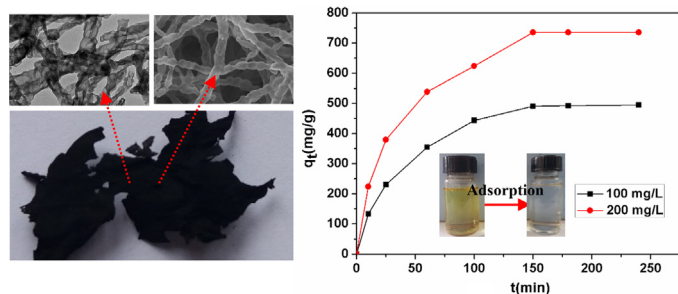
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GRAPHICAL ABSTRACT

Novel reusable bamboo-like polypyrrole nanofibrous mats were successfully prepared by non-emulsion electrospinning and in-situ polymerization. The adsorption capacity of Cr(VI) from aqueous solution was achieved as high as 961.5 mg/g at pH = 2. The flexible and integrity bamboo-like polypyrrole nanofibrous mats showed stable reusability and allowed easy handling during application.



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ABSTRACT

Development of highly efficient and easy-handling adsorbents for heavy metals removal is desirable for the remediation of our existing aquatic system. Herein, we demonstrated the novel bamboo-like polypyrrole nanofibrous mats for the removal of highly toxic hexavalent chromium (Cr(VI)) from aqueous solution. To achieve this target, the V_2O_5 nanofibrous templates were prepared via non-emulsion electrospinning technique and calcination, followed by in-situ polymerization of pyrrole. Benefiting from the special porous structure, high surface area and abundant adsorption active sites, the resulting bamboo-like polypyrrole nanofibrous mats exhibited a high Cr(VI) adsorption capacity up to 961.5 mg g^{-1} at room temperature, which can be well maintained for five adsorption/desorption cycles. The adsorption capacity for Cr(VI) can be enhanced with the decrease of pH and adsorption process belonged to the pseudo-second-order model. Furthermore, the adsorption isotherms of bamboo-like polypyrrole nanofibrous mats fitted the Langmuir isotherm model, and the adsorption mechanism of electrostatic attraction between bamboo-like polypyrrole nanofibers and Cr(VI) was presented. More importantly, the flexible and integrated bamboo-like polypyrrole nanofibrous membrane allowed easy handling during application, which is potentially used for heavy metal removal from aqueous solution.

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1. Introduction

The rapid development of modern industry has intrigued serious water pollution, which has greatly threatened the ecological environment and human health. Typically, heavy metal ions have become the major sources of water pollution due to massive discharge of wastewater from industries including cement ceramics, mining, battery manufacturing, electroplating, production of plastics, etc [1–3]. Among various heavy metal ions, Cr(VI) is extremely toxic and can be accumulated through food chain, thus causing huge damage to human body [4,5]. For instance, excessive intake of Cr(VI) can lead to cancers, nervous system failure, and kidney damage. Therefore, according to U.S. Environmental Protection Agency (US-EPA), the maximum allowable concentration of Cr(VI) in drinking water is 0.05 mg/L [6]. To meet this demand, development of efficient method for removing Cr(VI) before discharge to aquatic systems is urgent.

During the last decades, various techniques including ion exchange, electrochemical, chemical precipitation, and membrane filtration were developed for the removal of Cr(VI) from aqueous solution [7–10]. Although many achievements were obtained, most of these methods require high cost and/or high energy for their implementation. Besides, most of these methods are ineffective as the metal concentrations are lower than 20 mg/L. In contrast, adsorption has become the most versatile method due to its high-efficiency, low cost, flexibility and simplicity in operation [11]. In this concern, various adsorbents such as activated carbons [12], polymers [13,14], natural materials [15,16], clay minerals [17], and waste materials [18] were proposed. Especially, graphene and graphene oxide have shown the promising candidates as adsorbents for removal of Cr(VI) due to their large surface area and abundant active sites for adsorption [19,20]. To overcome the separation difficulty after adsorption, the magnetic graphene and graphene oxide were developed by introduction of magnetic nanoparticles [21–23]. However, some drawbacks including low adsorption capacity and easy oxidation/dissolution of iron nanoparticles (NPs) in acid solution restricted their further application for removal of Cr(VI). Therefore, it is still imperative to develop stable and easily available adsorbents with high adsorption capacity for Cr(VI) species from aqueous solution.

In recent years, polypyrrole (PPy) as one of the most popular conducting polymers has gained special attention in environmental remediation due to its easy synthesis, high environmental stability, and high adsorption efficiency [24–26]. Since it was reported for the Cr(VI) removal by Rajeshwar in 1993 [27,28], various PPy derivatives and composites were developed for removing Cr(VI) from aqueous media. It's been generally accepted that the large surface area of nanostructured PPy or PPy based composites is required for high adsorption efficiency of Cr(VI) in theory. Yao and coworkers [29] prepared the hierarchical porous PPy nanoclusters, which showed the higher adsorption capacity for Cr(VI) compared with traditional PPy nanoparticles because of larger surface area. To further enhance the adsorption capacity for Cr(VI), various nanostructured particles including Fe₃O₄ [30,31], silica [32], carbon nanotubes [33], and graphene derivatives [34] were used as ideal matrix for preparing PPy nanocomposites. Generally, the carbon nanotubes/PPy and graphene derivatives/PPy nanocomposites exhibited superior adsorption capacity compared with Fe₃O₄ and silica due to their large specific surface area and highly porous structures. However, there are two major challenges existing. On the one hand, the above carbon nanomaterials/PPy composites adsorbents are hard to be separated from aqueous media after adsorption. On the other hand, owing to the agglomeration of PPy particles, the dopant anions diffusion out of polymer is difficult, which would limit the adsorption capacity of adsorbents.

Consequently, easy-handling (easily separated from aqueous media after adsorption) PPy mats are of current interest in heavy metals removal [35,36]. Especially, superior surface area and highly porous structure endow the PPy mats with high adsorption performance for Cr(VI). For instance, Wang and coworkers [37] prepared PAN/PPy nanofibrous mats for the removal of Cr(VI), and the maximum adsorption capacity was 74.91 mg/g. Bhaumik and coworkers [36] prepared the PANI/PPy nanofiber, and the maximum Cr(VI) adsorption capacity reached 227 mg/g. Normally, the PPy mats are prepared by using the polymer fibers as templates. Therefore, adsorption efficiency is greatly limited to the physical property of templates. Furthermore, the structure stability is difficult to be guaranteed. Obviously, the adsorption efficiency of Cr(VI) is strongly depended on the structure and morphology of PPy nanofibers, which would intrinsically affect the surface area and adsorption active sites of nanofibers. Recently, our previous research demonstrated that easy-handling polypyrrole hollow nanofibrous mats with the high Cr(VI) adsorption capacity of 839.3 mg/g [38] could be achieved by combining emulsion electrospinning, calcination, and in-situ pyrrole polymerization. However, the easy aggregation of polystyrene@N, N-dimethylformamide emulsion within the solution required complicated steps to maintain the morphologies of precursors' fibrous mats. Therefore, easy preparation of polypyrrole nanofibrous mats with stable and special porous structure is quite important for Cr(VI) removal.

In this study, non-emulsion electrospinning and in-situ polymerization were combined to fabricate bamboo-like polypyrrole nanofibrous mats for the removal of hexavalent chromium (Cr(VI)) from aqueous solution. Benefiting from the special porous structure, high surface area and abundant adsorption active sites, as prepared bamboo-like polypyrrole nanofibrous mats exhibited superior adsorption capacity (961.5 mg g⁻¹) for Cr(VI) at room temperature. More importantly, the flexible and integrated bamboo-like polypyrrole nanofibrous mats showed stable reusability and allowed easy handling during application, which is potentially used for heavy metal removal from aqueous solution.

2. Experimental

2.1. Materials

Polyvinylpyrrolidone (PVP, Mw ~ 1,300,000), pyrrole, and vanadyl acetylacetonate were purchased from Sigma-Aldrich. Hydrogen chloride, potassium dichromate, sodium hydroxide, and N, N-dimethylformamide were obtained from Kelong Chemical Reagent Factory (Chengdu, China). All chemicals and agents were used without further purification.

2.2. Preparation of tubular V₂O₅ templates

Tubular V₂O₅ templates were prepared from electrospinning technique as shown in Fig. 1. Typically, 0.6629 g of vanadyl acetylacetonate was dissolved in N, N-dimethylformamide (4 mL) and stirred for 0.5 h. Then, 6 mL of EtOH were added into solution and stirred for 2 h. After that, 1.5 g PVP was dissolved into above solution and stirred for 12 h. The nanofiber was electrospun onto an aluminium foil with voltage of 15 kV. Meanwhile, the flow rate and distance between syringe needle tip and collector were set to be 20 cm and 1.0 mL/h, respectively. Finally, the tubular V₂O₅ templates were obtained by calcination according to the following procedure: room temperature to 330 °C, 5 °C/min and dwell for 2 h; 330–430 °C, 1 °C/min and dwell for 10 min.

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