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Radiation-induced graft polymerization for the preparation of a highly efficient UHMWPE fibrous adsorbent for Cr(VI) removal

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HIGHLIGHTS

- A high efficient fibers for Cr(VI) removal was prepared via radiation technique.
- Adsorption of Cr(VI) onto this fibrous adsorbent was followed pseudo-second-order.
- Maximum adsorbent capacity of Cr(VI) was found to be 295 mg/g.
- Cr(VI) was partially reduced to Cr(III) by the nitrogen species of adsorbent.

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ABSTRACT

A novel fibrous adsorbent containing amine and quaternary ammonium groups was prepared by radiation-induced graft of glycidyl methacrylate (GMA) onto ultra-high molecular weight polyethylene (UHMWPE) fiber and further modifying with triethylenetetramine (TETA) and glycidyl trimethylammonium chloride (GTA). The ATR-IR spectra and SEM observation demonstrated that amine and quaternary ammonium groups were immobilized onto the surface of UHMWPE fiber. The principal factors affecting the adsorption of Cr(VI) ions have been investigated including pH of the aqueous solution, contact time, temperature and coexisting anions. This novel fibrous adsorbent could effectively adsorb Cr(VI) in the range of pH 1–9, and the maximum adsorption capacity reached 295 mg/g at pH 3 and 25 °C based on the Langmuir isotherm. It was found that adsorption equilibrium could be achieved within 2 h for initial Cr(VI) of 100 mg/L, following the pseudo-second order model. The effect of coexisting anions (including SO_4^{2-}, $H_2PO_4^-$, NO_3^- and Cl^-) on the uptake of Cr(VI) was investigated in detail. Additionally, the adsorption saturated fiber could be regenerated by soaking in 0.5 mol/L NaOH solution, and the adsorption performance of this adsorbent could be maintained at 90% after eight cycles of adsorption-desorption. ATR-IR and XPS analysis revealed that Cr(VI) ions were adsorbed on the fiber adsorbent through ion exchange mechanism.

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1. Introduction

Contamination of water bodies by heavy metals present in industrial effluents has been attracting increasing attention over the past few decades. This contamination poses a serious threat to aquatic ecosystems and human health (Owlad et al., 2009). Chromium is one of the common toxic heavy metal ions found in surface and ground water, owing to its widespread use in steel production, leather tanning, electroplating, printing, and other industries (Setshedi et al., 2015). Chromium mainly exists in the environment in two oxidation states, as Cr (III), and Cr(VI), i.e., in the form of dichromate ($Cr_2O_7^{2-}$), hydrochromate ($HCrO_4^-$), or chromate (CrO_4^{2-}) in solution depending on pH (Qiu et al., 2014). Compared with Cr (III), Cr(VI) is more hazardous because it is a strong oxidizer and may cause epidermal irritation, vomiting, gastric ulceration, stomach cancer, and liver and kidney damage in human beings (Ren et al., 2014; Costa, 2003). Because of its high toxicity toward biological systems, the permissible level of Cr(VI) in industrial wastewater is 0.5 mg/L according to the Integrated Wastewater Discharge Standard of China (GB 8978–1996) (Wang et al., 2012). The World Health Organization has recommended a

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maximum acceptable Cr(VI) concentration of 0.05 mg/L in drinking water (Qiu et al., 2014). Thus, it is desirable to remove Cr(VI) from industrial sewage before discharge into the environment, in order to safeguard human health.

Current techniques used for Cr(VI) removal from aqueous solution and industrial effluents include reduction (Ke et al., 2011; Yoon et al., 2009), precipitation (Jafari et al., 2013), ion-exchange (Rengaraj et al., 2003; Tenorio and Espinosa, 2001), membrane separation (Modrzejewska and Kaminski, 1999; Piedra et al., 2015; Tapiero et al., 2015), electrocoagulation (Hamdan and El-Naas, 2014; Ölmez, 2009), and adsorption (Di Natale et al., 2015; Wang et al., 2013; Yu et al., 2013a). In comparison with other methods, adsorption has been more extensively studied because of its operational simplicity, cost-efficiency, and ability to recover Cr(VI) at a low concentration (Kong et al., 2014; Ma et al., 2014). A variety of adsorbents for Cr(VI) have been developed, including chitosan (Elwakeel, 2010; Sankararamakrishnan et al., 2006), silica (Liu et al., 2010; Zhao et al., 2011), resins (Tenorio and Espinosa, 2001), nanotubes (Tian et al., 2015), cellulose (Lin et al., 2013; Yang et al., 2014; Zhang et al., 2012), graphene (Wang et al., 2015), magnetic nanoparticles (Kara et al., 2015; Ni et al., 2015), and synthetic fibrous adsorbents (Wang et al., 2012; Kumar et al., 2008). Compared with granular and powder adsorbents, fibrous adsorbents have many advantages such as its relatively large surface area, rapid adsorption, easy regeneration and reusability, low cost, ease of use, and high mechanical stability (Deng and Bai, 2004; Yiğitoğlu and Arslan, 2005; Zhang et al., 2008). Therefore, it is of great practical and academic interest to develop effective fibrous adsorbents for removal of Cr(VI) from the aquatic environment.

Some fibrous adsorbents containing amine or quaternary ammonium groups have been prepared for the removal of chromium (Anirudhan et al., 2013; Kumar et al., 2008; Wang et al., 2011). Amine and quaternary ammonium groups on the fiber surface are considered to be the main sites for sorption through electrostatic interaction, ion exchange, or a combination of these two mechanism (Elwakeel, 2010). Furthermore, adsorbents containing quaternary ammonium groups can work efficiently over the entire pH range owing to the strong basic nature of the group (Toral et al., 2009), making these adsorbents appropriate for practical applications. In the traditional synthesis of quaternary ammonium adsorbents, a large amount of environmentally hazardous reagents such as chloromethyl methyl ether and chloroacetyl chloride is typically used to obtain a high quaternary ammonium group density (Huang et al., 2012; Kenawy et al., 2002; Zhang et al., 2008). However, these reagents are toxic, irritants, and carcinogens (Kong et al., 2014); hence, special equipment and devices are required during the adsorbent preparation. Therefore, a non-toxic method for the preparation of quaternary ammonium fibrous adsorbents is desirable.

Glycidyl methacrylate (GMA) has been grafted onto different matrix materials through radiation-induced graft polymerization (RIGP) (Han and Zhang, 2009; Takács et al., 2012; Ueki et al., 2012). As amine groups can be readily immobilized onto the GMA grafted substrates via a ring-opening reaction, functional groups such as primary amines, secondary amines, and tertiary amines can be added. These can be protonated to form $–NH_3^+$, $–NRH_2^+$, and $–NR_1R_2H^+$ in aqueous solution (Yu et al., 2013b) and may be further chemically modified to form quaternary ammonium groups (Anirudhan et al., 2013; Atia, 2006), which possess the ability to adsorb anion pollutants through electrostatic interaction or ion exchange. Compared to fibrous adsorbents containing only one kind of functional group (amine or quaternary ammonium), the novel fibrous adsorbent functionalized by both these groups would have higher adsorption capacity and can display efficient ion uptake under both acidic and alkaline conditions, which make more suitable for practical applications. In addition, the RIGP technique is suitable for pilot or industrial scale production. To the best of our knowledge, removal of Cr(VI) using such a fibrous adsorbent based on GMA via RIGP has not yet been reported.

In this work, a fibrous adsorbent carrying both amine and quaternary ammonium groups was prepared through radiation graft copolymerization of GMA and methyl acrylate (MA) onto ultra-high molecular weight polyethylene (UHMWPE) fiber. Subsequently, amination and functionalization with quaternary ammonium groups were performed. The fibrous adsorbent was characterized by attenuated total reflection infrared spectroscopy (ATR-IR) and scanning electron microscopy (SEM). Adsorption behavior of this fibrous adsorbent toward Cr(VI) ions was investigated in batch adsorption experiments to understand the effects of pH, adsorption kinetics, adsorption isotherms, competition with coexisting anions, and possible reusability. The relevant adsorption mechanism was proposed by analysis of the fibrous adsorbent before and after a Cr(VI) adsorption and regeneration test.

2. Experimental

2.1. Materials

UHMWPE fiber with a linear density of 3.6 denier, was purchased from Beijing Tongyizhong Special Fiber Technology & Development Co., Ltd. Glycidyl methacrylate purchased from Sigma Co. Ltd was used without further purification. Glycidyl trimethylammonium chloride (> 95%) was from Adamas Reagent Co. Ltd. Other reagents, purchased from Sinopharm Chemical Reagent Company, were of analytical grade. Nitrogen (99.99%) gas was purchased from Shanghai Louyang Gas Canned Co., Ltd. A stock solution of Cr(VI) was prepared by dissolving 2.8287 ± 0.0005 g of $K_2Cr_2O_7$ in 1000 mL of deionized water. $K_2Cr_2O_7$ was previously dried at 80 °C in a vacuum oven for 12 h. Working solutions were prepared by diluting Cr(VI) stock solutions to appropriate concentrations.

2.2. Preparation of UHMWPE fibrous adsorbent

The preparation process of UHMWPE fibrous adsorbent was illustrated in Scheme 1. The grafted UHMWPE fiber with epoxy groups was used in this study, which was synthesized by graft copolymerization of GMA and MA on UHMWPE fiber according to our method reported previously (Gao et al., 2015) (the sample was coded as UHMWPE-*g*-P(GMA-*co*-MA)).

The UHMWPE-*g*-P(GMA-*co*-MA) fiber was further modified with 50 vol% of triethylenetetramine (TETA) in 1,4-dioxane at 80 °C for 3 h under the atmosphere of N_2. The modified sample was washed several times with fresh ethanol and deionized water to remove the residual TETA, and then dried in a vacuum oven at 60 °C (referred by F1).

The TETA modified fiber (F1) was reacted with 10 wt% of glycidyl trimethylammonium chloride (GTA) in aqueous solution at 80 °C for 6 h under the protection of N_2. The resulting fiber (referred by F2) was washed several times with deionized water to remove the residual GTA and then dried at 60 °C in a vacuum oven.

2.3. Characterization of the modified fibers

Attenuated total reflection infrared spectrometry (ATR-IR) of the modified fiber was obtained using a Bruker Tensor 207 FT-IR spectrometer to analyze the functional groups on the fiber surface and recorded from 4000 cm^{-1} to 600 cm^{-1}. A scanning electron microscope (SEM, FEI Quanta-250) coupled with an energy dispersive spectrometer (EDS) was employed to observe the morphology of the fibers at an acceleration voltage of 10 kV. All

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