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Quaternized dimethylaminoethyl methacrylate strong base anion exchange fibers for As(V) adsorption



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HIGHLIGHTS

• QDMAEMA-g-PE/PP fibers have high adsorption capacity for As(V) ions.

• Adsorption of As(V) is independent on the solution pH over a wide range (4-10).

• As(V) adsorption rate of QDMAEMA-g-PE/PP fibers is considerably fast.

• The maximum adsorption capacity (q_{max}) was found to be 83.33 mg As(V)/g polymer.

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ABSTRACT

N,N-Dimethylaminoethyl methacrylate (DMAEMA) grafted polyethylene/polypropylene (PE/PP) nonwoven fibers (DMAEMA-g-PE/PP) was prepared by radiation-induced graft polymerization. DMAEMA graft chains on nonwoven fibers were quaternized with dimethyl sulfate solution for the preparation of strong base anion exchange fibers (QDMAEMA-g-PE/PP). Fiber structures were characterized by FTIR, XPS and SEM techniques. The effect of solution pH, contact time, initial As(V) ion concentration and coexisting ions on the As(V) adsorption capacity of the QDMAEMA-g-PE/PP fibers were investigated by performing batch adsorption experiments. The adsorption of As(V) by QDMAEMA-g-PE/PP fibers was found to be independent on solution pH in the range 4.00–10.00. Kinetic experiments show that the As(V) adsorption rate was rapid and As(V) adsorption follows pseudo second-order kinetic model. As(V) adsorption equilibrium data were analyzed using Langmuir and Freundlich adsorption isotherm model equations. Langmuir and Freundlich adsorption isotherm models fitted the experimental data well. The maximum adsorption capacity (q_{max}) calculated from Langmuir isotherm was found to be 83.33 mg As(V)/g polymer at pH 7.00. The adsorbent was used for three cycles without significant loss of adsorption capacity. The adsorbed As(V) ions were desorbed effectively by a 0.1 M NaOH solution.

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1. Introduction

Arsenic is one of the most toxic and ubiquitous element for human health and the environment (Chandra et al., 2010; Zhao et al., 2011a). Arsenic is mainly exists in two forms; arsenate, As (V) and arsenite, As (III) (Munoz et al., 2002). Arsenate or arsenite may become predominant depending on the oxidizing conditions in natural water. Arsenate is predominant in natural water under oxidizing conditions, while arsenite is predominant in natural water under nonoxidizing conditions. At about neutral pH, the predominant species are $H_2AsO_4^-$ and $HAsO_4^{2-}$ for arsenate, and uncharged H_3AsO_3 for arsenite. Arsenic is toxic and carcinogenic

http://dx.doi.org/10.1016/j.radphyschem.2014.04.011 0969-806X/© 2014 Elsevier Ltd. All rights reserved. element that exists primarily as oxyanion forms in water sources and has become a worldwide environmental issue. Arsenic removal from water sources and even drinking water systems has become very important for several years. According to chronic toxicological and carcinogenic effects of arsenic in drinking water, many authorities reduced the limit value. The World Health Organization (WHO) guideline value for arsenic in drinking water was provisionally reduced in 1993 from 50 to 10 µg/L (WHO, 1993). The limit value of arsenic in drinking water was also reduced to $10 \,\mu\text{g/L}$ by the European Community (EC) and the United States Environmental Protection Agency (USEPA). Therefore, the removal of arsenic from water sources is one of the most essential issues with regard to environmental protection and conservation. The major treatment technologies for the removal of arsenic include chemical precipitation, membrane filtration, adsorption and ion exchange processes (Camacho et al., 2009;

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Nguyen et al., 2009; Manning et al., 2002; Anirudhan and Unnithan, 2007). Among them, ion exchange and adsorption are the most widely studied treatment technologies due to the efficiency in aqueous solutions.

Recently, wide range of weak-base and strong-base anionexchange adsorbents that contain tertiary amine and guaternary ammonium groups attached to various adsorbent materials have been extensively studied for arsenic adsorption, such as cellulosic adsorbent modified with dimethylamine, dimethylaminoethyl methacrylate and quaternized triethylenetetramine (Anirudhan and Unnithan, 2007: Anirudhan and Jalaiamony, 2010: Tian et al., 2011: Anirudhan et al., 2012), glycidyl methacrylate grafted cotton cellulose modified with tetraethylenepentamine (Yu et al., 2013). glycidyl methacrylate resin functionalized with tetraethylenepentamine (Donia et al., 2011), polyethylene coated polypropylene grafted N-vinylformamide anion exchange fibers (Awual et al., 2008), ethylenediamine modified polyacrylonitrile ion-exchange fiber (Ruixia et al., 2002; Zhang et al., 2008a,b), polyaminesfunctionalized silica (Yoshitake et al., 2002, Fan et al., 2011), silica grafted with N,N-Dimethylaminoethyl methacrylate (Zhao et al., 2011b), ethylenediamine functionalized magnetic Fe₃O₄ core-shell nanorods (Babu et al., 2013) and commercial weak base Diaion resins (Diaion WA20 and Diaion WA30) (Awual et al., 2013).

N,N-Dimethylaminoethyl methacrylate (DMAEMA) is an acrylic monomer having tertiary amine functional groups. N,N-Dimethylaminoethyl methacrylate has been grafted onto many matrix materials such as synthetic polymers (e.g., polypropylene, polyethylene coated polypropylene), cellulose and silica in order to prepare functional adsorbents with specific property and used for the removal of anionic [As(V), Cr(VI), P(V) and N(V)] and cationic contaminants [Cu(II) and Hg (II)] from aqueous solutions (Burillo et al., 2013; Taleb et al., 2008; Akkaş Kavaklı et al., 2010; Tian et al., 2011: Li et al., 2012: Jiang et al., 2013: Oiu et al., 2009: Zhao et al., 2011b; Gao et al., 2010). Because DMAEMA carrying weak base such as tertiary amino functional groups that can be readily converted to positively charged quaternary ammonium groups having ion exchange properties, can be considered as a promising adsorbent material for the adsorption and separation of heavy metal ions. Polyethylene (PE), polypropylene (PP) and polyethylene coated polypropylene (PE/PP) fibers are important synthetic fibers that are commercially available and have been used as trunk materials after functionalization in many applications such as separation and water treatment (Akkaş Kavaklı et al., 2004a; Akkaş Kavaklı et al., 2004b; Shiraishi et al., 2003; Seko et al., 2004; Hegazy et al., 1999). Polyethylene/polypropylene (PE/PP) nonwoven fibers have lots of advantages as trunk materials; for instance, abundant, cheap, able to be applied in various flexibility forms such as fibers, nonwoven fabrics, threads, and cloths. However, polyethylene/polypropylene nonwoven fibers cannot be used as a plain adsorbent for the adsorption of target ions or molecules (contaminants or heavy metal ions) due to lack of functional groups. Therefore, a processing step is necessary for plain polyethylene/polypropylene nonwoven fibers which convert non-functional fiber surfaces into functional fiber surfaces by giving functional properties and making them reactive adsorbent for the interactions of target ions or molecules. Radiation induced graft polymerization process is one of the methods most frequently used to prepare functional fibrous polymeric materials (Nasef and Güven, 2012; Seko et al., 2005). Radiation graft polymerization offers an effective approach to introducing some desirable functional properties into the trunk polymer by selecting an appropriate hydrophilic functional monomer (acrylonitrile, acrylic acid, acrylamide, N,N-dimethylaminoethyl methacrylate, glycidyl methacrylate, methacrylic acid and N-vinyl-2-pyrrolidone) and optimizing the grafting conditions, thus tuning the grafting density of the trunk polymer and improving the adsorption affinity to target ions or molecules without affecting the backbone structure (Okamoto et al., 1985; Gupta and Chapiro, 1989; Gupta and Anjum, 2001; Akkaş Kavaklı et al., 2007a; Akkaş Kavaklı et al., 2007b; Mukherjee and Gupta, 1985; Liu et al., 2004).

In the present study, N,N-dimethylaminoethyl methacrylate strong base anion exchange fibers (QDMAEMA) for adsorption of As(V) was prepared by radiation induced graft polymerization of N,N-dimethylaminoethyl methacrylate onto the polyethylene/ polypropylene (PE/PP) nonwoven fibers and then DMAEMA grafted PE/PP fibers quaternized with dimethyl sulfate. Fiber structures were characterized by FTIR, XPS and SEM techniques. The effect of solution pH, contact time, initial As(V) ion concentration and coexisting ions on the adsorption capacity of the QDMAEMA-g-PE/PP fibers were investigated. Desorption of As (V) ions from the QDMAEMA-g-PE/PP fibers was examined using 0.1 M NaOH.

2. Experimental section

2.1. Materials and reagents

Polypropylene coated by polyethylene (PE/PP) nonwoven fibers were supplied by Kurashiki Seni Kako Co. Okayama, Japan. N,N-Dimethylaminoethyl methacrylate (DMAEMA) was purchased from Kanto Chemical, Tokyo, Japan and used without purification. Na₂HAsO₄.7H₂O, NaCl, KH₂PO₄ and K₂SO₄ was purchased from Sigma-Aldrich and NaNO₃ was purchased from Fluka. The arsenate stock solution (1000 mg As/L) was prepared from sodium salt heptahydrate (Na₂HASO₄·7H₂O) dissolved in ultrapure water. Other reagents were of analytical or higher grade and used as received. All solutions were prepared using ultrapure water (18.2 M Ω cm) produced from a TKAsmart2pure standard water dispenser (Thermo Electron LED, GmbH).

2.2. Preparation of DMAEMA grafted nonwoven fibers

Fiber adsorbent was prepared by radiation-induced graft polymerization of DMAEMA onto polyethylene/polypropylene (abbreviated as PE/PP fiber) nonwoven fiber. The PE/PP nonwoven fibers were sealed in polyethylene bags and purged with nitrogen gas. The PE/PP nonwoven fibers were irradiated by electron beam at a voltage of 2 MeV and a current of 3 mA. Irradiation was carried out in steel trays on a conveyor which transported the samples through the irradiation area, where the electron beam was directed downward. CTA (cellulose triacetate) film dosimeters were attached on the polyethylene bags during irradiation process. The polyethylene bags were positioned horizontally by placing on top of the dry ice $(-20 \degree C)$ in the tray at four different irradiation doses (30, 50, 100 and 200 kGy). After irradiation, the samples were stored at -80 °C until grafting process. The irradiated fibers were immersed in different concentrations of DMAEMA solutions ranging from 5% to 20% (w/w) in water at different temperatures. The detailed experimental conditions of DMAEMA grafting on PE/PP nonwoven fibers were given in our previous paper (Akkaş Kavaklı et al., 2007a). DMAEMA grafted nonwoven fibers hereafter abbreviated as DMAEMA-g-PE/PP.

2.3. Quaternization of the DMAEMA grafted fibers

Particular amount of DMAEMA grafted fibers were immersed in dimethyl sulfate solution in DMF. The mixture solution with fibers was heated in thermostated oil bath at 65 °C under nitrogen atmosphere for 24 h with stirring. The fibers were removed from reaction medium and then washed with DMF, water and methanol several times and dried under vacuum at room temperature Download English Version:

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