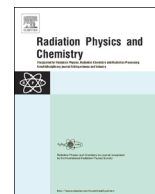




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Preparation and characterization of Fe(III)-loaded iminodiacetic acid modified GMA grafted nonwoven fabric adsorbent for anion adsorption



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HIGHLIGHTS

- A novel iron(III) loaded-iminodiacetic acid polymeric ligand exchanger was prepared.
- Iminodiacetic acid fibers have high iron(III) loading capacity.
- Iron(III) loaded-iminodiacetic acid fibers have affinity for phosphate.
- Novel adsorbent have high phosphate adsorption in acidic solutions.

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ABSTRACT

An Fe(III)-loaded chelating fabric with iminodiacetic acid (IDA) functional groups was prepared by radiation induced graft polymerization of an epoxy group containing monomer, glycidyl methacrylate, onto a nonwoven fabric made of polypropylene coated by polyethylene (PE/PP) and subsequent Fe(III) loading. Grafting conditions were optimized, and GMA grafted polymer was modified with iminodiacetic acid in isopropyl alcohol/water at 80 °C. In order to prepare the polymeric ligand exchanger (PLE) for the removal of phosphate, IDA fabrics were loaded with Fe(III) ions. Fe(III) loading capacity of IDA fabric was determined to be 2.83 mmol Fe(III)/g of polymer. For removal of phosphate anion, adsorption experiments were performed in batch mode at different pH (2–9) and phosphate concentrations. It was found that phosphate adsorption by the Fe(III)-loaded IDA fabric is maximum at pH 2.00. The effect of initial concentration of phosphate on the adsorption behaviour of Fe(III)-loaded IDA nonwoven fabric was determined at low phosphate concentrations (0.5–25 ppm) and at high phosphate concentrations (50–1000 ppm).

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1. Introduction

Polymers with specific functionalities can be obtained by either synthesizing new monomers carrying the special groups capable of interacting with the target metal ions and polymerizing them into practically applicable shapes and forms or by converting some groups on existing polymers or copolymers with suitable reactants into desired forms either by chemical or radiation treatment. Radiation induced graft polymerization (RGIP) is one of the processes for the manufacture of advanced polymeric materials by modifying widespread polymers (Nasef and Güven, 2012; Seko et al., 2005). Radiation grafted adsorbents have been used usually for separation and wastewater treatment (Akkaş Kavaklı et al., 2004a, 2004b; Shiraishi et al., 2003; Seko et al., 2004; Hegazy et al., 1999). Several types of ion exchange matrices have been

prepared by grafting monomers like acrylic acid, methacrylic acid, glycidyl methacrylate, dimethylaminoethylmethacrylate, acrylonitrile on trunk materials like polyethylene and polypropylene (Mukherjee and Gupta, 1985; Gupta and Chapiro, 1989; Akkaş Kavaklı et al., 2007a, 2007b; Kawai et al., 2000).

Phosphate is the major limiting nutrient in freshwater systems and excessive amounts of phosphate may cause eutrophication and pose significant environmental problems to water resources. The removal of phosphate from waters can be an effective method for the control of eutrophication in natural waters. Extensive studies on the effective treatment of phosphates in water systems have been previously conducted (Zhao and Sengupta, 1996, 1998, 2000; Boki and Tanada, 1987). The major removal technologies for the removal of phosphate include chemical precipitation, biological treatment, adsorption and ion exchange processes. Recently, adsorbents loaded with a transition metal having polymeric ligand exchanger (PLE) properties have been applied to remove phosphate (oxoanions or anionic contaminants) from water sources. A PLE consists of a polymer with chelating functional groups that

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can bind tightly to a transition metal, which can remove variety of ions from aqueous systems by complex formation. It is known that Cu(II), Fe(III), Al(III), Y(III), La(III), Mo(VI) Ti(IV) and Zr(IV) have specific affinity for oxoanions (Zhao and Sengupta, 2000; Chanda et al., 1988; Popat et al., 1994; Haron et al., 1995; Kanesato et al., 1988; Dambies et al., 2000; Balaji and Matsunage, 2002; Awual et al., 2011). In recent years, various transition metal-loaded synthetic or natural adsorbent materials have been developed and they have shown promising results in the removal of the hazardous anions from aqueous solutions. Anirudhan developed an iron(III)-coordinated amino-functionalised poly(glycidylmethacrylate)-grafted cellulose (Fe(III)-AM-PGMACell) and demonstrated that (Fe(III)-AM-PGMACell) can be used as an efficient adsorbent for the removal and recovery of phosphate from water and wastewater (Anirudhan and Senan, 2011). Unnithan et al. (2002) used iron(III)-loaded carboxylated polyacrylamide-grafted sawdust adsorbent for the removal of phosphate from water and wastewater. The iron(III)-loaded carboxylated polyacrylamide-grafted sawdust adsorbent exhibited a very high adsorption potential for phosphate ions. Zr(IV) and Fe(III)-loaded collagen fiber (denoted as Zr-ICF and Fe-ICF) adsorbents were prepared by Liao et al. (2006) used for the adsorption of phosphate in aqueous solutions. Wu developed a La(III)-loaded Chelex-100 resin for the removal of phosphate in wastewater. Adsorption of phosphate from water by La(III)-loaded Chelex-100 resin was investigated in the column mode. It was found that the La(III)-loaded Chelex-100 resin was able to remove phosphate efficiently from water, and the adsorption of phosphate was not affected by the presence of large amounts of anions such as chloride and sulfate (Wu et al., 2007). Zr(IV)-loaded phosphoric acid resin (RGP) prepared by Zhu and Jyo (2005) used to investigate its applicability in phosphate removal in the column mode. It was found that low concentration of phosphate could be removed at high flow rate due to high phosphate adsorption capacity of Zr(IV)-loaded RGP. Awual et al. (2011) developed an Zr(IV)-loaded bifunctional fibers containing both phosphonate and sulfonate for trace phosphate removal from water at high feed flow rate by ligand exchange fibrous adsorbent.

Although a number of polyvalent metal ions, including Cu(II), Fe(III), Zr(IV), Mo(VI) and Ti(IV), have been successfully immobilized on chelating adsorbents for oxoanions removal, the use of PLE nonwoven fabrics prepared by means of radiation induced graft polymerization has been very limited. A novel PLE nonwoven fabric having Fe(III)-loaded iminodiacetic acid functional groups selective for phosphate was prepared by means of radiation induced graft polymerization which is different from conventional anion exchange resins based on commercial crosslinked polystyrene matrices used for the removal of phosphate from aqueous solutions. Commercial adsorbents with iminodiacetic acid (IDA) functional group such as Chelex 100, Amberlite IRC 718, Purolyte S930, Lewatit TP 207 and Muromac A1 have been extensively studied for selective removal of heavy metals from wastewater and have also been used as a separation adsorbent in analytical chemistry for pre-concentration of trace metals from aqueous media due to the strong chelating properties of iminodiacetic acid functional group (Rahmi et al., 2007; Noureddine et al., 2008; Zainol and Nicol, 2009; Atia et al., 2008; Valverde et al., 2004; Vassileva and Furuta, 2003; Sumida et al., 2006). It is known that iminodiacetic acid functional group has high affinity for Fe(III) ions. In this paper, a study of phosphate adsorption using Fe(III)-loaded iminodiacetic acid nonwoven fabric as adsorbent is presented. The contact time of the phosphate adsorption was followed and the influence of pH on adsorption was determined in order to establish the optimum pH values for phosphate adsorption and to determine the capacity of Fe(III)-loaded iminodiacetic acid nonwoven fabric adsorbent. The outcome of this work could

also be useful for the further application of Fe(III)-loaded iminodiacetic acid nonwoven fabrics in the adsorption of trace contaminants such as arsenite, arsenate, chromate, selenite, selenate, thiocyanate and fluoride from waters.

2. Experimental

2.1. Materials

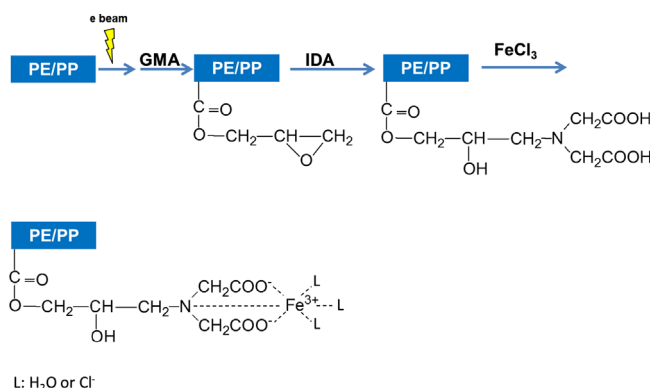
Nonwoven fabric polypropylene coated by polyethylene fabric was supplied by Kurashiki Sen-I Kako Co. Okayama, Japan. Methacrylic acid glycidyl ester was purchased from Tokyo Kasei Co., Japan. IDA was supplied Wako pure chemical industrial Ltd., Japan. Iron(III) chloride hexahydrate (FeCl₃·6H₂O), sodium chloride (NaCl) ACS reagent from Sigma-Aldrich, while sodium nitrite (NaNO₂), sodium nitrate (NaNO₃) were reagents from Merck and potassium dihydrogen phosphate (KH₂PO₄) was ACS reagent from Fluka. All chemicals used were of reagent grade. All solutions were prepared using ultrapure water (18.2 MΩ·cm) produced from a TKA Smart2Pure standard water dispenser (Thermo Electron LED GmbH, Germany).

2.2. Preparation of chelating IDA fabric

Scheme 1 shows the preparation of iminodiacetic acid containing nonwoven fabric. The preparation of nonwoven fabric grafted with side chains containing iminodiacetic acid (IDA) groups consisted of three steps; (1) grafting of an epoxy-group containing monomer, glycidyl methacrylate (GMA) by preirradiation grafting technique, (2) modification of epoxy group with iminodiacetic acid, and (3) preparation of Fe-IDA adsorbents by Fe(III) loading.

First, GMA was grafted onto PE coated PP nonwoven fabrics by pre-irradiation technique: irradiation by electron beam was performed in nitrogen atmosphere at ambient temperature using a cascade electron accelerator (Dynamitron, IEA-3000-25-2, Radiation Dynamics) at Takasaki Radiation Chemistry Research Establishment, Japan Atomic Energy Research Institute (JAERI). The concentration of GMA solution was set at 10 (w/w) % in methanol as the solvent. Preparation of GMA grafted nonwoven fabric was given in our previous paper in details (Akkaş Kavaklı et al., 2007a, b).

Second, the GMA grafted nonwoven fabric was immersed in 0.425 M iminodiacetic acid in isopropyl alcohol/water (20/80 (V/V) solution. The reaction was performed at 80 °C for different time periods for different degree of GMA grafted samples to construct the conversion curve. After binding of iminodiacetic acid groups onto the epoxy group of grafted polymer branches, the remaining unreacted epoxide groups were hydrolyzed with acid solution for 2 h at 80 °C. Subsequently nonwoven fabric was washed with



Scheme 1. Fe(III)-loaded iminodiacetic acid modified GMA grafted nonwoven fabric.

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