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Preparation and adsorption property of xylan/poly(acrylic acid) magnetic nanocomposite hydrogel adsorbent

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ABSTRACT

Adsorbents based on natural polysaccharides have attracted increasing interest because of their low-cost and biodegradability, particularly, polysaccharide-based nanocomposite adsorbents. In this study the xylan/poly(acrylic acid) magnetic nanocomposite hydrogel adsorbent was prepared from wheat straw xylan and Fe_3O_4 nanoparticles, and its adsorption property was studied on methylene blue removal. The prepared hydrogel adsorbent had a semi-interpenetrating network structure and exhibited a macroporous structure with interconnected porous channels. Super-paramagnetic characteristic behavior was observed from magnetic analysis using a vibrating sample magnetometer. The optimum condition for methylene blue adsorption on the adsorbent was found at pH 8 with an adsorbent dosage of 3 g/L and an initial concentration of 400 mg/L, and the removal percentage reached above 90%. The adsorption isotherm of methylene blue on the prepared hydrogel adsorbent was fitted to the Langmuir model, and the pseudo-second-order kinetic model could describe the adsorption process. All obtained results indicated that the prepared hydrogel adsorbent is promising for water treatment applications.

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1. Introduction

Wastewater pollution from paper making, cosmetic, electroplating, smelting and dyeing industries is a serious environment problem (Mittal, Gajbe, & Mittal, 2008; Sui et al., 2012; Ai & Jiang, 2012). Removals of hazardous materials from wastewater have been studied by many researchers. According to previous studies (Liu, Wu, Chiu, Suen, & Chu, 2007; Selcuk, 2005; Kornaros & Lyberators, 2006; Shen, Shen, Wen, Wang, & Liu, 2011), various treatment methods have been used to remove hazardous materials, such as ion exchange, chemical precipitation, membrane filtration and adsorption. Adsorption of hazardous materials on adsorbent is considered to the most effective method for wastewater treatment in terms of cost, ease of operation, flexibility and simplicity of design (Crini & Badot, 2008). Recently, there has been increased interest on adsorbents based on natural polymers because of their low-cost and biodegradability. Natural polymers contain abundant functional groups which are very useful for removing hazardous materials. Cellulose, chitosan and cyclodextrin based adsorbents have been reported and used in the areas, such as drug delivery (Luo, Liu, Zhou, & Zhang, 2009), immobilization of enzyme (Luo & Zhang, 2010) and water treatment (Shen et al., 2011; Luo & Zhang, 2009; Huang, Liu, Zhang, Xu, & Hu, 2012).

Xylan is an abundant polymer in nature, and it was found in hardwood and straw, comprising roughly one-fourth to one-third of most plant materials. Xylan consists mainly of β-1→4 linked xylose unites with other substituents arranged in different proportions, and it has excellent hydrophilicity, biodegradability, and biocompatibility (Ebringerová, Hromadkova, & Heinze, 2005; Oliveira et al., 2010); therefore, xylan has become a very promising raw material for preparing novel materials. We have studied the chemical structure and separation of straw xylan (Sun, Fowler, Rajaratnam, & Zhang, 2010; Sun, Jing, Fowler, Wu, & Rajaratnam, 2011), and straw xylan-based hydrogels with excellent swelling and response properties had been prepared (Sun, Jing, & Wang, 2013a; Sun, Wang, Jing, & Mohanathas, 2013b), and the hydrogels can be applied in medical field and wastewater treatment. However, there are few reports that focused on xylan-based nanocomposite hydrogel.

In our previous paper (Sun, Jing, Wang, & Liu, 2014), physical–chemical properties of xylan/poly(acrylic acid) magnetic nanocomposite hydrogel were studied, and the hydrogel had excellent thermal stability, magnetic- and pH-sensitive properties. The thermal stability of the hydrogels enhanced because of an increase in the contents of xylan and Fe_3O_4 nanoparticles; however, the equilibrium swelling ratio decreased with increasing the

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contents of Fe_3O_4 nanoparticles and xylan. This paper investigated the adsorption property of the hydrogel on methylene blue (MB) removal, and the effects of solution pH, adsorbent dosage, initial MB concentration and contact time on MB removal were discussed. The adsorption mechanism was also studied using adsorption isotherm and kinetics models.

2. Experimental

2.1. Materials and reagents

Xylan was prepared according to a previous paper (Sun et al., 2013b) with some modifications in which the hemicellulosic material was further treated using 0.05 mol/L HCl solution at 50 °C at a 1:20 (g/mL) solid to liquid ratio to obtain pure xylan, and the xylan material contained 93% xylose and 5% arabinose. Acrylic acid was purchased from the Tianjin Kermel Chemical Reagent Company in China. *N*,*N*′-Methylenebisacrylamide, ammonium persulfate and anhydrous sodium sulfite were purchased from the Tianjing Hongyan Chemical Reagent Factory in China. $FeCl_3{\cdot}6H_2O$, $FeSO_4{\cdot}7H_2O$ and MB were provided by the Tianjing Tianli Chemical Reagent Factory in China. All other reagents used were of analytical grade.

2.2. Preparation of hydrogel adsorbent

The Fe_3O_4 nanoparticles were prepared using chemical co-precipitation method. $FeCl_3{\cdot}6H_2O$ and $FeSO_4{\cdot}7H_2O$ (mol ratio = 2:1) were dissolved in 150 mL of distilled water, and the solution was transferred into a three neck flask filling using N_2. The pH value of the mixed solution was adjusted to 10.0 using $NH_3{\cdot}H_2O$, and this solution was stirred at 70 °C for 1 h. After the reaction, the black substance was removed using a magnet, and the obtained product was washed using distilled water and ethanol alternately for five times. After drying for 24 h in an oven, the black substance was ground to particles of 120 nm diameter, and Fe_3O_4 nanoparticles were finally prepared for further use.

0.4 g of xylan powder was dissolved in the mixed solution of 9 mL distilled water and 1 mL acrylic acid. 0.02 g of *N*,*N*-methylenebisacrylamide and 0.28 g of Fe_3O_4 nanoparticles were added into the above solution, and the mixture was stirred using mechanical agitation under ultrasonic irradiation. Next, 0.01 g of $(NH_4)_2S_2O_8$ and 0.01 g of $NaSO_3$ were added into the mixture solution. After the reaction finished, the black hydrogel materials were removed and cut into uniform size pieces (0.5 × 0.5 × 0.5 cm), and then, these pieces were soaked in distilled water for 48 h. During this period, it was necessary to change the water regularly for washing away unreacted materials. The prepared hydrogel adsorbent were pre-frozen at −20 °C and then freeze-dried at −50 °C for 48 h. Finally, the xylan/poly(acrylic acid) magnetic hydorgel adsorbent was prepared.

2.3. Characterization of the prepared adsorbent

FT-IR spectroscopy was used to analyze the chemical structure of the prepared hydrogel adsorbent. The ground hydrogel samples were mixed with KBr and then pressed into discs that were analyzed using a FT-IR spectrometer (Nicolet 510) in the range from 4000 to 500 cm^{-1}. The magnetic property of the prepared adsorbent was evaluated using a vibrating sample magnetometer (VSM, Lakeshore 7307), and the hysteresis loop was obtained in a magnetic field varying from −1.5 to +1.5 T. The morphology of the adsorbent was observed on a scanning electron microscope (SEM, SUPRA 55), and the hydrogel samples that swollen in distilled water and the solutions of different MB concentrations were first freeze-dried, and the morphology of the samples was observed and photographed.

The mechanical property of the prepared hydrogels with and without Fe_3O_4 nanoparticles was analyzed. Dynamic mechanical analysis (DMA, TA instrument Q800 179 series) was used to determine the compressive modulus of the swollen hydrogel samples. To reach swelling equilibrium, hydrogels were incubated in distilled water for 24 h at room temperature before test. The disk shaped samples were 1 cm × 0.5 cm (diameter × height) in dimension and were tested in compression mode at 25 °C.

2.4. Adsorption experiment

To evaluate the adsorption property of the prepared adsorbent, MB was chosen to remove from aqueous solution using the adsorbent. The effects of solution pH, adsorbent dosage, and initial MB concentration on the adsorption performance of the prepared adsorbent were tested. Varied numbers of the hydrogel adsorbent sample were added in 10 mL solutions of MB, and the pH values of the MB solutions were adjusted using 0.1 mol/L HCl or NaOH solution, and the solutions were sealed at 25 °C for 48 h. The MB concentrations of the remaining solutions were measured using UV–vis spectrometer at its maximum wavelength of 665 nm. The equilibrium adsorption amount of MB, q_e (mg/g), was calculated using the following equation:

$$q_e = \frac{(C_o - C_e)V}{W} \quad (1)$$

Removal percentage (%) was calculated using the equation:

$$\text{Removal percentage } (\%) = \frac{C_o - C_e}{C_o} \times 100\% \quad (2)$$

where C_o (mg/L) and C_e (mg/L) are the MB concentrations at initial and adsorption equilibrium states, respectively; V (L) represents the volume of MB solution; W (g) is the weight of the adsorbent.

The effect of contact time on MB adsorption was performed as follows: 0.5 g of the prepared hydrogel adsorbent was immersed in a 20 mL solution of MB. The pH value of the MB solution was adjusted using 0.1 mol/L HCl and 0.1 mol/L NaOH. After the certain time intervals, the concentration of MB solution was determined using above method. Adsorption amount of MB, q_t (mg/g), at time t (h) was calculated using the following equation:

$$q_t = \frac{(C_o - C_t)V}{W} \quad (3)$$

where C_t (mg/L) is the MB concentration at adsorption time t (h).

3. Results and discussion

3.1. Chemical structure of the prepared adsorbent

Fig. 1 displays the FT-IR spectra of xylan, hydrogel adsorbent and Fe_3O_4 nanoparticles. The broad band at 3400 cm^{-1} was attributed to the O-H stretching vibration, and the absorption peak at 2930 cm^{-1} originated from the C-H stretching vibration. The FT-IR spectrum of xylan presented a band at 1630 cm^{-1} that arose from the water adsorption, and the sharp absorption peak at 1044 cm^{-1} was assigned to the C—O and C—C stretching and the glycosidic linkage V(C—O—C) contributions, which is the characteristic absorption of xylan (Sun et al., 2013b). The band at 562 cm^{-1} appeared in the FT-IR spectrum of Fe_3O_4, which is related to the vibration of the Fe—O (Luo et al., 2009). However, the Fe—O characteristic peak in the FT-IR spectrum of the prepared adsorbent was shifted to lower wavenumber, which may be a result of physical effect. The band at 1726 cm^{-1} appeared in the FT-IR spectrum of the prepared adsorbent, which arises from carbonyl groups of poly(acrylic acid) (Wang

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