Preparation of Hemicellulose-based Hydrogel and its Application as an Adsorbent Towards Heavy Metal Ions

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Water contamination by heavy metal ions is a worldwide problem. In this work, a type of hydrogel, based on hemicellulose, was prepared *via* graft copolymerization. The reaction time and cost were reduced by modifying the traditional preparation process. The hemicellulose-based hydrogel was an opaque, smooth, well-distributed, and high resilience hydrogel. To better understand the application of hemicellulose-based hydrogel as an absorbent for the removal of Pb(II) (lead nitrate) from heavy metal contaminated water, the hydrogel was used to adsorb Pb(II) from aqueous solution. The various factors that influenced the Pb(II) removal efficiency were studied, including concentration and temperature. The results showed that the hemicellulose-based hydrogel was assessed as an ideal absorbent towards Pb(II), showing maximum monolayer adsorption capacity of 5.88 mg/g. The Pb(II) adsorption isotherm was determined to further understand the adsorption mechanism. The results demonstrated that the adsorption of the hemicellulose-based hydrogel on Pb(II) belongs to the monolayer adsorption and that adsorption conforms to the Langmuir model better than the other model. It revealed that the adsorption process of Pb(II) on hemicellulose-based hydrogel was spontaneous and endothermic in nature. The hemicellulose-based hydrogel was proven as a good material for potential application.

Keywords: Hemicellulose; Hydrogel; Adsorption; Heavy metal ions; Kinetic

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INTRODUCTION

Hemicelluloses are a type of hetero-polysaccharides that are present, together with cellulose and lignin, in the primary and secondary cell wall of wood and annual plants (Yang *et al.* 2011; Cheng *et al.* 2015). They are considered to be the second most abundant polysaccharide in nature after cellulose, with an estimated production of 60 billion tons per year (Timell 1967; Gatenholm and Tenkanen 2003). Because natural polysaccharides are non-toxic, biocompatible, biodegradable, and renewable, they have been widely applied in different fields (Crini 2005; Mizrahy and Peer 2012; Yang *et al.* 2015). In general, polymers from renewable resources can be categorized into three groups (Yu *et al.* 2006). The first group is the natural polymers, such as polysaccharides and proteins that exist in nature in their polymeric form and can be extracted directly from biomass. The second group consists of synthetic polymers such as polylactic acid (PLA) synthesized by using monomers extracted from nature. The third group includes polymers that are synthesized by microorganisms through fermentation of renewable monomers. Hemicelluloses from renewable resources have gained increasing importance in recent years as fossil fuels are becoming less available (Lindblad 2010; Sun *et al.* 2013).

Hydrogels made from biomaterials have attracted increasing attention in biology-related applications since the 1960s due to their ability to retain large amounts of water within their structure (Peppas *et al.* 1985). They have exhibited potential application in many fields such as agriculture (Chu *et al.* 2010), hygienic products (Kamat and Malkani 2003), biotechnology (Sannino *et al.* 2000), drug delivery (Voepel *et al.* 2009), environmental protection (Güçlü *et al.* 2010), and numerous other applications.

Heavy metal pollution is a global environmental problem that causes serious threats to human health and ecosystem (Hua *et al.* 2012; Prucek *et al.* 2015; Xia *et al.* 2015). Pb(II) is a commonly present non-degradable environmental contaminant that can enter the body through the food chain, soil, water, and air, and cause great harm to the body's cells. Recently, various methods for the removal of heavy metal ions from wastewater have been extensively investigated. These technologies include chemical precipitation, ion-exchange, adsorption, membrane filtration, coagulation–flocculation, flotation, and electrochemical methods. Comparatively speaking, adsorption of inorganic non-metallic materials was recognized as an effective and economic method, such as activated carbon and diatomite. Adsorption is regarded as one of the most economically favorable and technically easy methods that have been employed for the removal of Pb(II) from water (Hena 2010). Nowadays, hydrogels have attracted particular attention for removing heavy metal ions from waste water (Wu and Li 2013). Due to the porous structure of hydrogels, water and solutes diffuse through the matrix, including metal ions (Dorkoosh *et al.* 2002a,b). This work mainly discusses the preparation and the properties of hemicellulose-based hydrogel and their application in treatment of wastewater containing heavy metal ions. Langmuir and Freundlich isotherms were used to analyze the adsorption data, as well as recovery tests of hydrogels and metal ions.

EXPERIMENTAL

Materials

Hemicellulose used in this work was extracted from hardwood hydrolysate by ethanol (Asia Symbol Pulp and Paper Co., Ltd., Rizhao, China). All of the chemicals and reagents used for the experiment and analysis were analytical grade (Sigma Chemical Reagent Co., Ltd., St. Louis, MO, USA). Acrylic acid (AA) was purified prior to use, whereas the others were used without further purification. The morphology features and surface characteristics of the hydrogel samples were observed using scanning electron microscopy (JEOL, JSM-6460LV, Japan).

Methods

Preparation of hydrogel-based hemicellulose

The synthesis process is shown in Fig. 1. The hemicellulose was dispersed in distilled water with ultrasonic vibrations for 10 min. Then the hemicellulose dispersion liquid was mixed with emulsifier. The hemicellulose dispersion and the acrylamide were added to the above partial neutralized AA solution by magnetic stirring for 30 min at room temperature. The crosslinking agent was added to the above mixed solution by magnetic stirring for 10 min. Potassium persulfate and sodium bicarbonate were dissolved in distilled water and then added as an initiator system under the magnetic stirring for 10 min. Then the reaction was processed for 1 to 2 h without stirring. The hydrogel products were carefully washed thoroughly with deionized water and then dried to a constant mass at 40 °C in a vacuum drier.

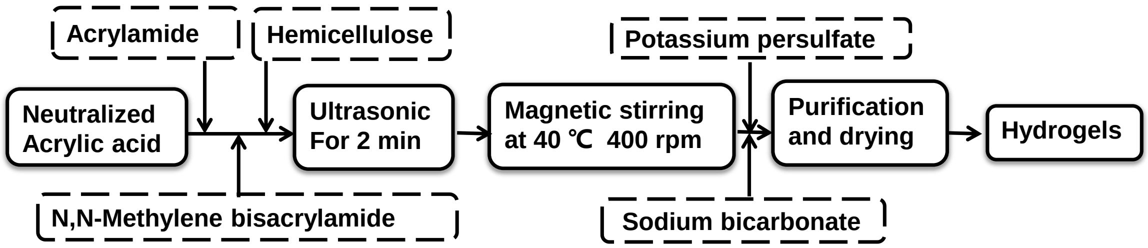

Fig. 1. Polymerization process of hemicellulose-based hydrogels

The preparation of hemicellulose-based hydrogels was optimized by an orthogonal experiment. When evaluating the product by the degree of swelling, the factors were: amount of AA, initiator content, and neutralization degree. The factors and levels are shown in Table 1.

Table 1. Factors and Levels of the Synthesis Conditions

Level	Factor		
	m(AA):m(HC)	Initiator (wt.%)	Neutralization Degree of AA (%)
1	15:1	1.5	80
2	10:1	1.0	70
3	5:1	0.5	60

The hemicellulose dosage was 5.0 wt.%, the acrylamide (AAm) amount was 25 wt.%, and the weight of the crosslinker was 0.1 wt.%.

Water absorbency measurement

The hydrogel sample was immersed in distilled water for a certain time then it was taken out and the excess water was removed by a 100-mesh gauze. The weight of the swollen samples were measured. The liquid absorbency (Q, g/g) was calculated by the following equation Eq. 1,

$$Q = \frac{M_2 - M_1}{M_1} \quad (1)$$

where M_2 (g) and M_1 (g) are the swollen and dry weights of the samples, respectively.

Pb(II) adsorption measurement

The standard curve of the $Pb(NO_3)_2$ (lead nitrate) solution was drawn by a UV spectrophotometer (Geneys 10S, Thermo Electron Co., Ltd., Waltham, MA, USA) at the maximum absorption wavelength. The $Pb(NO_3)_2$ solution concentrations were 1 ppm, 2 ppm, 5 ppm, 10 ppm, and 20 ppm. The isolated adsorbent (0.01 g) was added to 20 mL concentration gradient of $Pb(NO_3)_2$ solution, was shaken at room temperature for 6 h, and then the concentration of Pb(II) in the solution was measured. The adsorbing capacity (*Q*; mg/g) was calculated using Eq. 2,

$$Q = \frac{V \cdot (C_o - C_e)}{W} \tag{2}$$

where C_o (mg/L) and C_e (mg/L) are the origination and ending of adsorption of metal ion concentration, respectively, W (g) is dry weight of adsorbent, and V (L) is the volume of solution.

Isotherm studies were conducted with different concentrations at 30 °C, 40 °C, and 50 °C.

RESULTS AND DISCUSSION

Optimization of the Hemicellulose-based Hydrogel

The swelling degree of the synthetic products was investigated as the index in the three factors and three levels of the orthogonal test. The results are shown in Table 2.

Table 2. Range Analysis of Hemicellulose-based Hydrogel Synthesis Conditions

No.	m(AA):m(HC)	Initiator (wt.%)	Neutralization (%)	Error	Swelling Degree (g/g)
1	1	1	1	1	356
2	1	2	2	2	426
3	1	3	3	3	565
4	2	1	2	3	410
5	2	2	3	1	446
6	2	3	1	2	603
7	3	1	3	2	387
8	3	2	1	3	398
9	3	3	2	1	473
X_{1j}	1347	1153	1432	1200	-
X_{2j}	1459	1345	1234	1416	-
X_{3j}	1258	1566	1398	1448	-
R	201	413	198	248	-

The range analysis results showed that the influence factors on the swelling degree were initiator > m(AA):m(HC) > neutralization. The optimum level was as follows: initiator was 0.5 wt.%, m(AA):m(HC) was 10:1, neutralization degree of AA was 80%. Under the optimized condition mentioned above, the swelling degree of the hydrogel reached 603 g/g.

The analysis of variance results are shown in Table 3. These results revealed that the initiator was significant, and m(AA):m(HC) was less significant while the neutralization degree was non-significant.

For the above reason, the neutralization was insignificant. When the neutralization degree was 60%, the swelling degree obtained better data than range analysis. At the same time, the neutralization degree was reduced, production costs were also reduced, as well as secondary pollution from sodium hydroxide. Therefore, the neutralization degree chosen was 60%.

Table 3. Variance Analysis of Hemicellulose-based Hydrogel Synthesis Conditions

	R^2	DF	F	F(0.10)	Significance
m(AA):m(HC)	6762.89	2	2.82	4.32	
Initiator (wt.%)	43274.89	2	18.01	4.32	**
Neutralization (%)	1322.89	2	0.55	4.32	
Error	4804.44	4			

Based on the analysis, the hemicellulose-based hydrogel optimal synthesis conditions were as follows, temperature 40 °C, hemicellulose (HC) amount 5.0 wt.%, AAm amount 25 wt.%, the amount of crosslinking agent MBA 0.1 wt.%, the amount of initiator 0.5 wt.%, m(AA):m(HC) = 10:1, and the neutralization degree of AA of 60%. Under the preparation condition mentioned above, the swelling degree of the hemicellulose-based hydrogel reached 603 g/g.

Characterization of Hydrogel

Based on the optimum synthesis conditions, the hemicellulose-based hydrogels were obtained. The images of hydrogel samples are shown in Fig. 2.

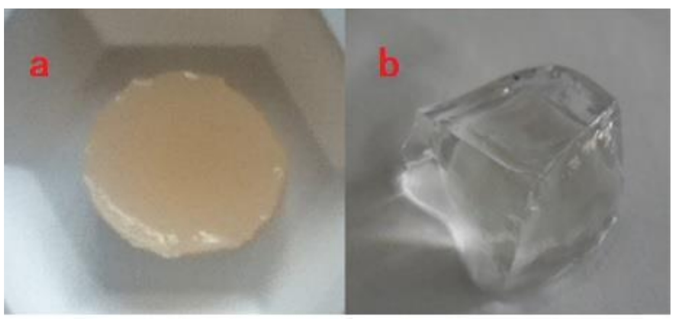

Fig. 2. Hemicellulose-based hydrogel before (a) and after (b) swelling

The hemicellulose-based hydrogel before (Fig. 2a) swelling was an opaque, smooth, well-distributed, and highly resilient hydrogel. After water absorption, the sample became transparent and colorless without disintegration, and had gained some mechanical strength.

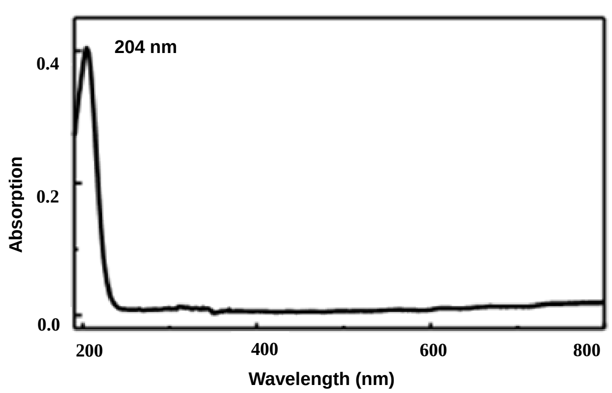

Fig. 3. Full wavelength scanning of $Pb(NO_3)_2$

Hemicellulose-based Hydrogel On Pb(II) Adsorption Performance

The $Pb(NO_3)_2$ solution was scanned by a UV spectrophotometer at the maximum absorption wavelength to draw the standard curve (Fig. 3). The maximum absorption wavelength was 204 nm.

The standard curve achieved under the different concentrations of $Pb(NO_3)_2$ is shown in Fig. 4.

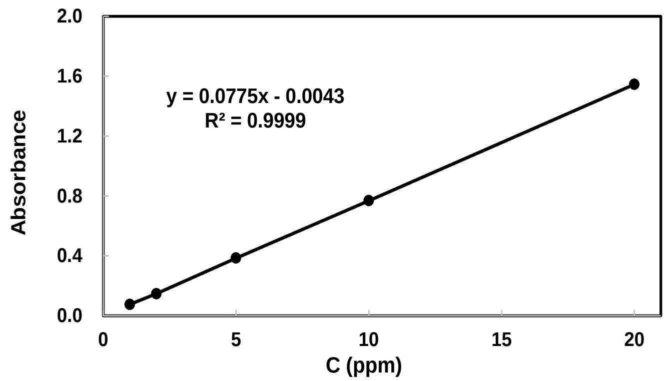

Fig. 4. The standard curve of solution $Pb(NO_3)_2$

Figure 4 illustrates the standard curve of the $Pb(NO_3)_2$ solution. The absorbance value (*A*) is shown by Eq. 3,

$$A = 0.0775X - 0.0043 \tag{3}$$

where *X* (ppm) is the concentration of $Pb(NO_3)_2$ solution, *A* is the absorbance value, the R^2 value was 0.9999, which indicates an excellent performance.

Hemicellulose-based hydrogels at various temperatures in different concentrations of $Pb(NO_3)_2$ solution equilibrium adsorption quantity (Q_e/mg·g^{-1}) are shown in Table 4.

Table 4. Equilibrium Pb(II) Adsorption (Q_e/mg·g^{-1}) onto Hemicellulose-based Hydrogels

Concentration (ppm)	Temperature (°C)		
	30	40	50
2	1.04	1.25	1.55
5	2.62	3.17	3.62
10	3.53	3.75	4.11
20	4.21	4.65	5.08

Figure 5 illustrates the adsorption isotherms of the hemicellulose-based hydrogel. With increased temperature, the adsorption capacity of hemicellulose-based hydrogel for $Pb(NO_3)_2$ solution increased, indicating that the elevated temperature was favorable for adsorption.

The Langmuir and Freundlich equation was used to fit the adsorption parameters of hemicellulose-based hydrogel on Pb(II). The fitting method referred to the adsorption isotherm of hydrogel on Pb(II) (Shawabkeh and Tutunji 2003).

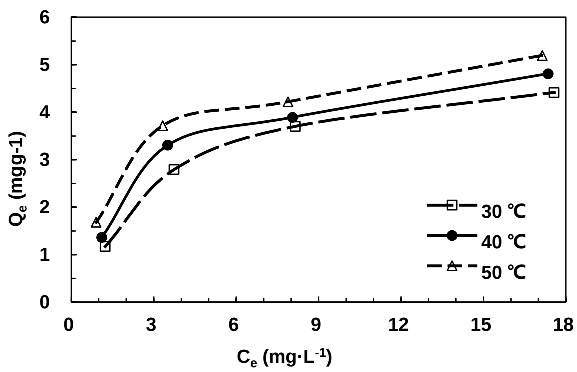

Fig. 5. Adsorption isotherm curves of Pb(II) onto hemicellulose-based hydrogel

The Langmuir isotherm is also known as a "single-molecule adsorption theory"; its equation is as follows:

$$\frac{1}{Q_e} = \frac{1}{Q_{max}} + \frac{1}{Q_{max} \cdot K_l} \cdot \frac{1}{Q_e} \quad (4)$$

The Freundlich isotherm is based on an empirically derived relationship. Its equation is as follows,

$$\ln Q_e = \ln K_f + \frac{1}{n} \ln C_e \quad (5)$$

where Q_e (mg/g) is the equilibrium absorption capacity of hemicellulose-based hydrogel and Q_{max} (mg/g) is the maximum adsorption amount. When a monolayer adsorption unit *m* is the mass of hemicellulose-based hydrogel, C_e (mg/L) is the concentration of $Pb(NO_3)_2$ solution; when adsorption is equilibrium, K_l is the Langmuir adsorption constant relatively, K_f and $1/n$ are the Freundlich adsorption constants relatively.

According to Eqs. 4 and 5, the plotting of the C_e/Q_e-C_e fitting the Langmuir adsorption and the $\ln Q_e$-$\ln C_e$ fitting the Freundlich adsorption is shown in Figs. 6 and 7, respectively. According to Figs. 6 and 7 of the Langmuir and Freundlich adsorption isothermal regression equation under different temperatures, *Kl*, *Kf*, and 1/*n* were calculated by Eqs. 4 and 5. The results are shown in Table 5.

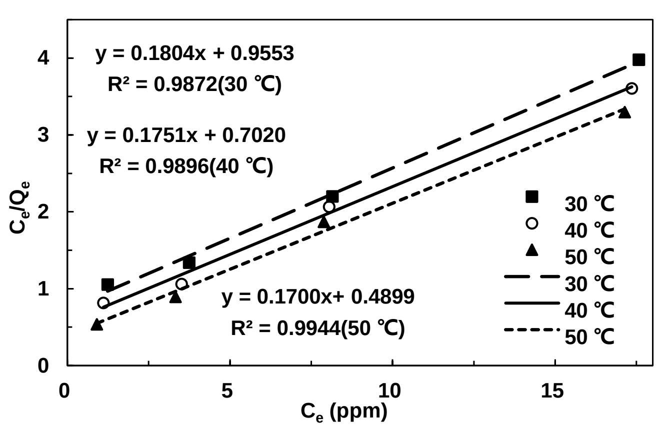

Fig. 6. Langmuir equilibrium isotherm of Pb(II) adsorbed onto hemicellulose-based hydrogel

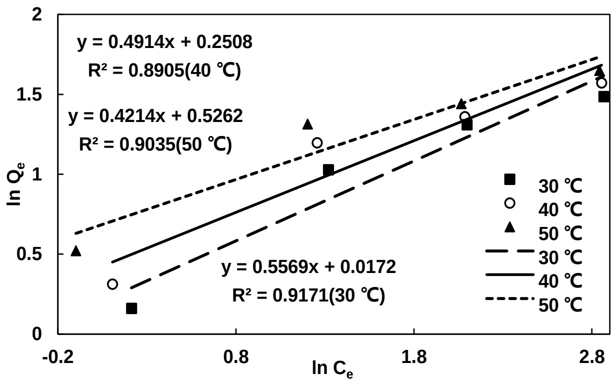

Fig. 7. Freundlich equilibrium isotherm of Pb(II) adsorbed onto hemicellulose-based hydrogel

Table 5. Thermodynamic Parameters for Adsorption of Pb(II) onto Hemicellulose Based Hydrogel

T (°C)	Langmuir			Freundlich		
	Q_{max} (mg·g^{-1})	K_l	R^2	K_f	1/*n*	R^2
30	5.54	0.1890	0.9872	0.9829	0.56	0.9171
40	5.71	0.2495	0.9896	1.2928	0.49	0.8905
50	5.88	0.3471	0.9944	1.6925	0.42	0.9035

As shown in Fig. 6 and Table 5, an increased temperature increased the Q_{max} as well as the adsorption capacity of hemicellulose-based hydrogel for $Pb(NO_3)_2$ solution, which indicated that the elevated temperature was beneficial for adsorption. This was consistent with the results achieved above in Fig. 5. When the temperature was 50 °C, Q_{max} reached the maximum of 5.88 mg/g, which showed that hemicellulose-based hydrogel had a good adsorption performance on Pb(II).

As shown in Fig. 7 and Table 5, 1/*n* was 0.56, 0.49, and 0.42, when the temperature was 30 °C, 40 °C, and 50 °C, respectively. This illustrated that hemicellulose-based hydrogel had a good adsorption performance on Pb(II).

The Langmuir and Freundlich models indicated that hemicellulose-based hydrogel had a good adsorption performance on Pb(II). The Langmuir adsorption constant was higher than the Freundlich adsorption constant, at the same time, Langmuir adsorption constant was greater than 0.98. Concerning the adsorption of hemicellulose-based hydrogel on Pb(II), the Langmuir adsorption isothermal equation was better. The results demonstrated that the adsorption of hemicellulose-based hydrogel on Pb(II) belongs to the monolayer adsorption and adsorption conformed to the Langmuir model better than the other one.

Analysis of Hemicellulose-based Hydrogel Sample

Figure 8 shows the morphology of the hydrogel sample. Part a shows an SEM image with 2000 times enlargement, whereas part b is a 5000 times enlargement. The results showed that the swelling properties were mainly determined by the hydrogels microstructure, such as multi-fold structure, uniform undulant surface, and specific surface area. As shown, the sample appeared to have a smooth and undulant surface.

Some sunken domains appeared in the surface. The undulant surface was convenient for the penetration of water into the hydrogel, while the rapid adsorption of hemicellulose-based hydrogels quickly reached swelling equilibrium. The sunken domains and the undulant surface was suitable absorption of Pb(II). The hydrogels prepared in this work were assessed as ideal absorbents for Pb(II) and novel materials with potential application prospects.

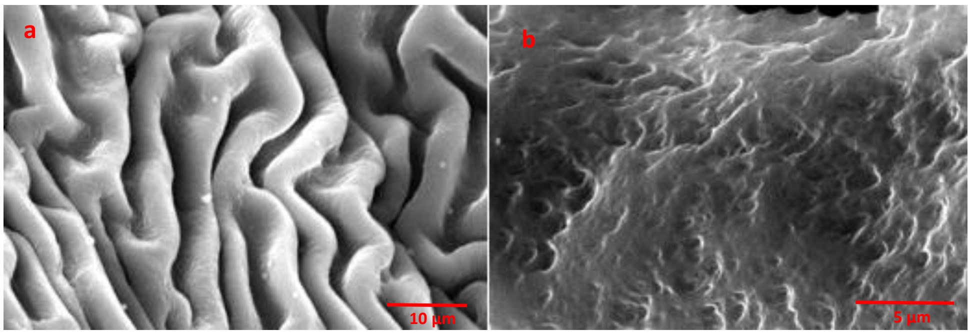

Fig. 8. SEM images of hemicellulose-based hydrogel sample

CONCLUSIONS

1. The synthesis conditions of hemicellulose-based hydrogel were optimized, where m (AA): m (HC) = 10:1, and the weight ratio (to monomers) of HC and AAm was 5.0% and 25%, respectively. The neutralization degree of AA was 60%, weight ratio (based on monomers) of the cross-linker MBA and the initiator was 0.1% and 0.5%, respectively. Under the optimal conditions, the hemicellulose-based hydrogel sample attained the maximum swelling degree of 603 g/g.
2. The hemicellulose-based hydrogel was assessed as an ideal absorbent toward Pb(II), showing a maximum monolayer adsorption capacity of 5.88 mg/g.
3. The Langmuir adsorption constant was greater than 0.98, which exceeded the Freundlich adsorption constant. Therefore, the Langmuir adsorption isothermal is the better equation for measuring hydrogels.
4. The adsorption of hemicellulose-based hydrogel on Pb(II) belongs to the monolayer adsorption, and the adsorption conforms to the Langmuir model better than the Freundlich model. The hemicellulose-based hydrogel is indeed a type of material with potential application prospect.

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