



Article Adsorption of Metallic Ions on Amidoxime-Chitosan/ Cellulose Hydrogels

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Abstract: Adsorption using natural compounds is an attractive separation technique for recovering heavy metals from aqueous media. Although chitosan, which is a natural polysaccharide, is an environmentally benign adsorbent, it dissolves in an acidic aqueous medium. In this study, we prepared adsorbents consisting of chitosan modified with amidoxime groups for improving metal adsorptivity, and cellulose for improving gel stability using an ionic liquid, and examined their adsorption characteristics for metal ions. The prepared amidoxime-chitosan/cellulose hydrogels had a mechanical strength without cross-linking. All the investigated metals were adsorbed on the amidoxime-chitosan/cellulose hydrogels in the following adsorptivity order: Cu \approx Ag > Ni > Zn. The adsorptivity of the metal ions increased with pH due to a proton exchange reaction. From the Langmuir adsorption isotherm, the Langmuir constant for Cu exceeded those of other metals because amidoxime has higher Cu affinity. The pseudo-second-order reaction model best described the adsorption kinetics with metal chelate formation being the rate-determining step. Because amidoxime-chitosan/cellulose hydrogels had higher physical stability and higher Cu selectivity, they were found to be a promising, environmentally benign adsorbent.

Keywords: chitosan; adsorption; metal ion; ionic liquid

1. Introduction

Heavy metals, dyes, phenols, detergents, and pesticides are common water pollutants. Since toxic heavy metals are likely to bioaccumulate through the food chain, heavy metals must be removed from aqueous media. Several separation techniques, including coagulation, adsorption, membrane filtration, and electrochemical methods have been studied. Among them, adsorption is very attractive, since it is a low cost and efficient technique for the remediation of heavy metals from aqueous media [1]. Different types of adsorbents, natural or synthetic, have been proposed for metal removal. Polysaccharides like cellulose, chitosan, starch, and alginate, which are abundant in nature, are typical natural adsorbents [2]. The use of polysaccharide homopolymers for removing metals is cost-effective. Moreover, polysaccharide homopolymers were easily converted to more adsorptive forms by chemical modification, blends, and composites of polysaccharides and copolymers [2].

Chitosan, which consists of D-glucosamine with β -(1,4) bonds, obtained by the deacetylation of chitin that is generally contained in crustacean shells, shows excellent adsorption capacity of metal ions due to its abundant amino groups and is expected to be an environmentally friendly tool for the removal of metal ions from wastewater [3]. In addition, the adsorption capacity and selectivity can be easily improved by the chemical modification of the highly reactive amino groups in chitosan [3]. However, chitosan itself is easily soluble in acidic solutions due to the protonation of amino groups. Cross-linking is generally used to make it insoluble, although cross-linking agents are usually not biocompatible. As a means of avoiding chemical cross-linking, the cross-linking of



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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). chitosan by gamma and electron beam irradiation has been proposed [4,5]. However, this method requires special equipment. Recently, it was reported that complex polysaccharide hydrogels of chitosan and cellulose, prepared from ionic liquids, such as 1-butyl-3-methyl imidazolium chloride and 1-ethyl-3-methyl imidazolium acetate ([Emim][Ac]), were stable in acidic solutions without cross-linking treatment and had an excellent adsorption capacity of nickel and copper ions [6,7]. Although the LiOH/urea/H₂O system can also dissolve polysaccharides such as chitosan and cellulose [8], ionic liquids are more environmentally friendly than LiOH, which is a highly corrosive chemical, because ionic liquids are green corrosion inhibitors [9].

Amidoxime groups have shown the high selectivity for strontium [10], copper [11,12], gallium [13,14], vanadium [15], and uranium [16–19]. We previously reported that crosslinked chitosan modified with amidoxime groups improved the adsorption capacity and selectivity for copper ions more than cross-linked chitosan [20]. In this study, complex polysaccharide hydrogels of chitosan modified with amidoxime groups and cellulose were prepared using [Emim][Ac] and their adsorption characteristics for metal ions were examined.

2. Materials and Methods

2.1. Materials

In this study, we used cellulose powder (type A; Advantec, Toyo Roshi Kaisha, Ltd., Tokyo, Japan) and chitosan (50–200 [cps], 100% deacetylated, Katokichi, Kan-nonji, Japan). The ionic liquid used was 1-ethyl-3-methyl imidazolium acetate ([Emim][Ac]) (Kanto Chemical Co. Inc., Tokyo, Japan). Metal nitrates were used. All the remaining reagents were of analytical grade and used without further purification. Amidoxime chitosan was prepared according to the same procedure (Scheme 1) described in our previous paper [20]. To identify the cyanoethyl chitosan and amidoxime chitosan, the FT-IR measurements (IRAffinity-1, MIRacle 10, Shimadzu Co., Kyoto, Japan) of modified chitosan were performed by an ATR method.



Scheme 1. Preparation of amidoxime chitosan.

2.2. Preparation of Amidoxime-Chitosan/Cellulose Hydrogels

Cellulose and amidoxime-chitosan were added to [Emim][Ac] and stirred at 80 °C for 48 h to form a homogeneous solution with respective concentrations of 5% (w/w). The obtained homogeneous solution was dropped into 200 mL of ion exchange water using a 1-mL syringe and left to stand for 1 h to allow the hydrogels to harden. Then, the hydrogels were collected, washed thoroughly with ion-exchanged water, stored in ion-exchanged water, and filtered just before use.

2.3. Adsorption of Metal Ions on Hydrogels

Adsorption tests of various metal ions, including Cu(II), Ni(II), Zn(II), and Ag (I) on hydrogel beads were measured to evaluate the adsorption ability by shaking 0.3 g of gel beads in 25 mL of the test solution that contained 1 mmol/dm³ of the individual metal ions at 100 rpm for 48 h at 30 °C. The pH values were adjusted using the following solution: 0.1 mol/dm³ HCl/CH3COOH/CH3COONa. After attaining equilibrium, the mixtures were filtered to separate the gel beads.

The adsorption isotherms were measured to evaluate the maximum adsorption capacity of both adsorbents by shaking 0.2 g of the gel beads with 25 mL of the test solutions containing individual metals that ranged from 0.25 to 30 mmol/dm³ at pH 6. The initial and residual concentrations of the metal ions in the filtrate were measured by inductively coupled plasma atomic emission spectrometry (ICPS-8100, Shimadzu, Japan). The pH values of the solution were measured with a pH meter (F-52, Horiba, Japan) before and after the adsorption experiment. The amount of adsorbed metal ions was calculated from the difference in the metal concentration between the initial solution and the filtrate. We calculated the amount of metal adsorbed on the hydrogel [q (mmol/g-gel)] with the following equation:

$$q = \frac{C_o - C_e}{M} \times V \tag{1}$$

where C_o and C_e are the metal concentrations [mmol/dm³] before and after adsorption. *V* is the volume of the solution [dm³], and *M* is the weight of the hydrogel [g].

3. Results and Discussion

3.1. Characteristics of Amidoxime-Chitosan/Cellulose Hydrogels

The FT-IR spectra of chitosan, cyanoethyl chitosan, and amidoxime chitosan are shown in Figure 1. The characteristic adsorption band of the nitrile group appears at around 2250 cm^{-1} [21] in the IR spectrum of cyanoethyl chitosan, and in the IR spectrum of amidoxime chitosan this peak appeared. From the elemental analysis, the degree of substitution of the amidoxime group is estimated as 0.12.



Figure 1. FT-IR spectrum of chitosan, cyanoethyl chitosan, and amidoxime chitosan.

Figure 2A,B shows photo images of the chitosan/cellulose and amidoxime chitosan/ cellulose hydrogels after copper ion adsorption. The chitosan/cellulose hydrogel was uniformly blue, and the amidoxime-chitosan/cellulose hydrogels were green, suggesting that both hydrogels had different complexation mechanisms with copper ion. Both hydrogels were stored in ion-exchanged water for three months and photo images are shown in Figure 2C,D. The surface of the chitosan/cellulose hydrogels (D) were disintegrated, and the amidoxime-chitosan/cellulose hydrogels (C) retained their spherical shape. The ability



of the hydrogen bonding acceptor of amines was weaker than that of the amidoximes [22]. Compared to chitosan, this state increased the interaction with the hydroxy groups in cellulose, resulting in greater mechanical strength.

Figure 2. Images of chitosan or amidoxime-chitosan/cellulose gel beads after copper adsorption, immediately after copper adsorption ((**A**) chitosan, (**B**) amidoxime chitosan), and after three months ((**C**) amidoxime chitosan, (**D**) chitosan).

3.2. Effect of Contact Time on Adsorption

Since the adsorption of metal ions on hydrogels involves the diffusion of metal ions in gels, we examined the time needed for equilibrium adsorption. The time dependency of the adsorption was investigated by contacting 0.2 g of hydrogels with 16.7 mL of acetate buffer solution (pH 6) that contained 1 mmol/dm³ copper ions at 100 rpm at 30 °C. Figure 3 shows the relationship between the contact time and the amount of copper ions adsorbed. The adsorption equilibrium was reached in 24 h. Hereafter, contact time was 48 h to be safe.

3.3. Effect of Initial pH on Adsorption

The pH of the solution is a key parameter that influences the adsorption of the metal ions Cu, Ni, Zn, and Ag on the hydrogels. Figure 4 shows the effect of the initial pH on the adsorption of the metal ions. All the metals investigated were adsorbed on the hydrogels in the following adsorptivity order: Cu \approx Ag > Ni > Zn. Although metal selectivity obeyed the Irving–William series (Ni < Cu > Zn), this trend did not correspond to that of the chitosan/cellulose hydrogels (Cu > Zn > Ni) [7]; it corresponded to that of the cross-linked amidoxime-chitosan resins [20]. As described in our previous paper [7], cellulose does not have the adsorption ability of metal ions, and the metal adsorption and metal selectivity was attributed to the amidoxime-chitosan. The metal ions form chelates, which are coordinated by the nitrogen atoms of the primary amino groups and the oxygen atoms of the oxime groups and are adsorbed by the cation exchange with the protons of the oxime groups. Therefore, the adsorptivity of metal ions increased with pH.



Figure 3. Effect of contact time on copper adsorption on amidoxime-chitosan hydrogels (gel dosage: 0.2 g, agitation speed: 100 rpm, temperature: 30 °C).



Figure 4. Effect of initial pH on metal adsorption on amidoxime-chitosan hydrogels. (Gel dosage: 0.3 g, agitation speed: 100 rpm, temperature: 30 °C).

3.4. Adsorption Isotherm

Figure 5 shows the adsorption isotherms of the metal ions on the hydrogels at pH 6. The isotherm data were analyzed with the Langmuir, Freundlich, and Temkin models [23]. The equations of the three models are expressed, respectively, in:

$$q = \frac{q_{max}K_LC_e}{1 + K_LC_e} \tag{2}$$

$$\mathbf{q} = K_F C_a^{1/n} \tag{3}$$

$$q = B_T \ln K_T + B_T \ln C_e \tag{4}$$

where q_e and K_L in Equation (2) are the maximum amount of adsorption, K_F and 1/n are the Freundlich constant and adsorption intensity in Equation (3), and B_T and K_T represent the heat of adsorption associated with Temkin's constant and the Temkin isotherm constant.

Parameters were determined by a non-linear regression program included in Sigmaplot 14. The parameters obtained are listed in Table 1, with the Langmuir constants with chitosan/cellulose hydrogels [7]. The Langmuir isotherm model generally corelated better with the adsorption process of metals than the other models by considering R^2 values. It was reported that the Langmuir model well explained the adsorption behaviors of metals on chitosan/cellulose composites [6-8], suggesting that metal ions chemically interact with gels. The solid lines in Figure 5 were calculated by Langmuir parameters. The calculated lines approximately agreed with the experimental data. From Table 1, the Langmuir constant for Cu is larger than those of other metals, suggesting that amidoxime has a higher affinity for Cu [11,23]. This corresponds to that of cross-linked amidoxime chitosan [20]. In chitosan/cellulose hydrogels, the Langmuir constants were generally smaller than those of the amidoxime-chitosan/cellulose hydrogels, and that for Cu was lower than those of Ni and Zn [7] suggesting that amidoxime enhanced Cu adsorptivity. The sharp rise of the isotherm curve of Cu means that this hydrogel can efficiently recover copper ions of low concentration. Adsorption capacities of copper on chitosan or chitosan composites were reported to be within 0.7~3 mmol/g-resin [2]. Maximum adsorption capacities of this hydrogels were lower than those of previous studies. Although one of the reasons is that cellulose has no metal adsorptivity, we consider further the effect of gel composition on gel stability and adsorptivity in the next stage.



Figure 5. The adsorption isotherm of metal on amidoxime-chitosan hydrogels (gel dosage: 0.3 g, pH: 6, agitation speed: 100 rpm, temperature: 30 °C).

Table 1. Adsorption isotherm parameters.

	Langmuir				Freundlich			Temkin		
Metal	<i>q_{max}</i> [mmol/g]	<i>K_L</i> [dm ³ /mmol]	<i>K_L</i> [dm ³ /mmol] [5]	R^2	1/n	K _F	R^2	K_T	B_T	<i>R</i> ²
Cu	0.152	3.48	0.0121	0.879	0.185	0.0952	0.962	36.5	0.0199	0.886
Ag	0.156	0.554		0.970	0.258	0.0677	0.959	43.3	0.0209	0.964
Ni	0.092	0.097	0.162	0.932	0.513	0.0127	0.885	39.6	0.0127	0.801
Zn	0.046	0.102	0.171	0.850	0.505	0.007	0.829	157	0.00392	0.723

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3.5. Adsorption Kinetics

Adsorption kinetics are one of the main factors for applying the adsorption process. We examined this using three representative kinetic models for chemisorption: pseudo-firstorder, pseudo-second-order, and intraparticle diffusion [24]. The recommended linearized equation of the three models is expressed with the following equations, respectively [24];

$$\ln(q_e - q_t) = \ln q_e - k_1 t \tag{5}$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \tag{6}$$

$$q_t = k_{IP} t^{0.5} + C_{IP} \tag{7}$$

where q_e and q_t are the amount of metal adsorbed on the hydrogel at equilibrium and time t, respectively; k_1 , k_2 and k_{IP} are the rate constants of pseudo-first-order reactions, pseudo-second-order reactions, and interparticle diffusion. C_{IP} is the boundary layer thickness.

The parameters determined by a non-linear regression program included in Sigmaplot 14 are listed in Table 2. In Table 2, q_e (exp) represents the average value of the steady state of q in Figure 3. The q_e (calc) value evaluated by the pseudo-second-order model shows good agreement with the q_e (exp) value, while the q_e (calc) value evaluated by the pseudo-first-order model was smaller than the q_e (exp) value, suggesting that the pseudo-second-order kinetic model well matched the experimental data. This is also supported by the R^2 values. The regression line based on the interparticle diffusion model (Equation (7)) does not pass through the origin and this suggests different mechanisms control the adsorption process [24]. Therefore, it is inferred that chemisorption is the rate-determining step in the adsorption process.

Kinetic Model	Parameters						
Pseudo-first-order	$k_1 (h^{-1})$ 0.206	<i>q_e</i> (exp) 0.0565	<i>q_e</i> (calc) 0.0458	<i>R</i> ² 0.901			
Pseudo-second-order	<i>k</i> ₁ (g/(mmol h)) 8.97	<i>q_e</i> (exp) 0.0565	<i>q_e</i> (calc) 0.0552	<i>R</i> ² 0.998			
Intraparticle	k _{IP} (mmol/(g h ^{0.5})) 0.0166	<i>C_{IP}</i> (mmol/g) 0.0034		R ² 0.885			

Table 2. Kinetic parameters and coefficients of determination.

Figure 6 shows the plot of the relation of Equation (6) using experimental data in Figure 3. A linear regression of measurement and calculated values yielded an R^2 of 0.998, suggesting that the two active sites available on the hydrogels coordinate with copper ions to form the chelate, and this step is rate-determining.



Figure 6. Plot of the pseudo-second-order model based on Equation (2) using data shown in Figure 2.

4. Conclusions

In this study, we prepared adsorbents consisting of chitosan modified with amidoxime groups and cellulose using an ionic liquid, [Emim][Ac], without cross-linking and examined their adsorption characteristics for metal ions. The amidoxime-chitosan/cellulose hydrogels were more stable than the chitosan/cellulose hydrogels. Metal ions were adsorbed on the amidoxime-chitosan/cellulose hydrogels in the following adsorptivity order: $Cu \approx Ag > Ni > Zn$. The adsorptivity of the metal ions increased with pH. From the Langmuir adsorption isotherm, the Langmuir constant for Cu exceeded those of other metals because of a higher Cu affinity with the amidoxime group. The pseudo-second-order reaction model best described the adsorption kinetics, suggesting that metal chelate formation is a rate-determining step. The amidoxime-chitosan/cellulose hydrogels had higher physical stability and higher Cu selectivity.

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