



Studies on the interaction of mercury(II) and uranyl(II) with modified chitosan resins

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Abstract

Chitosan polymer was cross-linked using glutardialdehyde. The resin obtained was chemically modified through the reaction with ethylenediamine and (3-amino-1,2,4-triazole-5-thiol) to produce chitosan/amine and chitosan/azole resins, respectively. The resins obtained were tested for the uptake of Hg^{2+} and UO_2^{2+} from their aqueous solutions. The chitosan/amine resin showed higher uptake towards Hg^{2+} and UO_2^{2+} compared to chitosan/azole. This was attributed to the fact that the azole moiety does not furnish efficient attachment to the metal ions compared to the amine active sites which are more compact. Selective separation of Hg^{2+} from UO_2^{2+} was achieved at $\text{pH} < 2$. This was explained by the ability of Hg^{2+} to adsorb on chitosan resin through ion-exchange in the acidic media while UO_2^{2+} would not. Uptake values up to 2.0 and 1.7 mmol/g were reported for Hg^{2+} and UO_2^{2+} , respectively, on chitosan/amine. Application of the Langmuir model to the adsorption data indicated a higher binding of Hg^{2+} to the resin compared to UO_2^{2+} . The adsorbed Hg^{2+} or UO_2^{2+} on the studied resins was eluted using H_2SO_4 and HCl, respectively.

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1. Introduction

The metal ions produced in wastes are of great environmental and economical interest. Usual treatments for extraction of metal ions from effluents consist of flotation, solvent extraction, precipitation, ion-exchange and electrochemical separation (Ajiwe et al., 2000). These methods sometimes suffer from

problems such as excessive time requirements, high costs and production of highly toxic sludges. As an alternative to the above methods, sorption techniques have been applied using activated carbon, clays, fly ash, biomass, etc. (Dambies et al., 2000; Malakul et al., 1998; Filho et al., 1995; Absalan et al., 2003). Research efforts have been directed at developing economical and efficient sorbents to eliminate mercury (Jeon et al., 2003; Kawamura et al., 1998a) and uranium (Egawa et al., 1988; Pekel et al., 2000; Zhang et al., 2003) from effluents.

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Chitosan as a material of biological origin has recently been used for recovery of metal ions (Lee et al., 2001; Varma et al., 2004; Arrascue et al., 2003; Cárdenas et al., 2001; Kawamura et al., 1997, 1998b). This biopolymer is produced by *N*-deacetylation of chitin which is extracted from several sources such as arthropods and fungal biomass (Dambies et al., 2001). Chitosan has many useful features such as hydrophilicity, biocompatibility, biodegradability and antibacterial property. Since it is harmless to humans, chitosan is widely used in food and pharmaceutical processes, medical and agricultural drugs. Chitosan is also used as an effective coagulant aid in drinking water treatment (Kawamura et al., 1993).

In the present work chitosan was cross-linked with glutardialdehyde and subsequently treated with epichlorohydrine and ethylene diamine or 3-amino-1,2,4-triazole-5-thiol. The uptake behaviour of the resins obtained towards Hg^{2+} and UO_2^{2+} was investigated.

2. Experimental

2.1. Chemicals

Chitosan used in this study was Aldrich product of FW 161, Brookfield viscosity 800,000 cps, CAS 9012-76-4, Batch #15403BB. Glutardialdehyde, epichlorohydrine and ethylenediamine (en) were Aldrich products. 3-amino-1,2,4-triazole-5-thiol (azole) was obtained from Organica Ltd., Germany. All other chemicals were Prolabo products and were used as received. Mercuric chloride and uranyl chloride were used as the source for Hg^{2+} and UO_2^{2+} , respectively.

2.2. Preparation of chitosan beads

Chitosan (5 g) (31 mmol of monomeric units) was dissolved in 2.0% aqueous acetic acid (250 ml). The chitosan solution was dropped into 0.1 M NaOH solution where a gelatinous precipitate of chitosan was formed, filtered off then washed thoroughly with distilled water (Becker et al., 2000).

2.3. Cross-linking of chitosan with glutardialdehyde

The gelatinous precipitate obtained in the previous step (31 mmol of monomeric units) was suspended in

100 ml methanol (Becker et al., 2000). A 1.19 ml (3.1 mmol) of 25% aqueous solution of glutardialdehyde was added then stirred at room temperature for 3 h followed by heating at 70 °C for 4 h. Afterwards, the product was isolated by filtration and washed with ethanol followed by distilled water and labeled as “CR”.

2.4. Reaction of chitosan resin with epichlorohydrine

The cross-linked chitosan particles obtained in the previous step were suspended in 70 ml isopropyl alcohol to which 5 ml epichlorohydrine (62.5 mmol) dissolved in 100 ml acetone/water mixture (1:1 v/v) was added. The contents were stirred for 24 h at 60 °C. The solid obtained was filtered off and washed several times with ethanol followed by water and labeled as “CR-Cl”.

2.5. Reaction of chitosan resin with ethylenediamine

Five grams of “CR-Cl” was suspended in 100 ml ethanol/water mixture (1:1 v/v) then ethylenediamine (5 ml) was added. The reaction mixture was stirred at 60 °C for 12 h then the product obtained was washed with ethanol followed by water. The chitosan-amine produced was dried in air and labeled as “CR-amine”.

2.6. Reaction of chitosan resin with (3-amino-1,2,4-triazole-5-thiol)

Five grams of “CR-Cl” was suspended in 100 ml of ethanol/water mixture (1:1 v/v) and 3 g of (3-amino-1,2,4-triazole-5-thiol) was added then the reaction mixture was heated at 70 °C for 12 h while stirring. The chitosan-azole produced was washed with ethanol followed by water then dried in air and labeled as “CR-azole”.

2.7. Estimation of the active sites on the obtained chitosan resins

The concentration of the amine active sites in the obtained resins was estimated using the volumetric method (Latha et al., 1991). A 20-ml HCl (0.05 N) solution was added to 0.1 g resin and conditioned for 15 h on a shaker. The residual concentration of HCl

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