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A Highly Efficient Chelating Polymer for the Adsorption of Uranyl and Vanadyl Ions at Low Concentrations

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Abstract. A new polymer containing double amidoxime groups per repeating unit was synthesized to enhance the metal ion uptake capacity. The adsorption properties of this new polymeric adsorbent, amidoximated poly(N,N′-dipropionitrile acrylamide), for U(VI), V(V), Cu(II), Co(II) and Ni(II) ions were investigated by batch and flow-through processes at very low concentration levels (ppb). The chelating polymer showed high adsorption capacity for uranyl as well as vanadyl ions. In selectivity studies from a mixture of metal ions in aqueous solutions, the adsorbent showed high selectivity for uranyl and vanadyl ions in the following order: U(VI) $>$ V(V) $\gg$ Co(II) $=$ Cu(II) $\gg$ Ni(II) as determined by calculating the distribution coefficients D, of corresponding ions. The adsorption of uranyl and vanadyl ions from natural seawater by the new adsorbent was also examined in flow through mode.

Keywords: chelating polymer, poly(dipropionitrile acrylamide), amidoximation, uranyl, vanadyl ion adsorption

Introduction

The design, synthesis and application of polymer supported reagents having chelating polymers that can selectively complex metal ions from multi-component solutions have been an important area of research during the last decades (Collega et al., 1980; Lutfor et al., 2000; Trochimezuk et al., 2001; Maria et al., 2001; Zhanhai et al., 2002). Chelating polymers are effective adsorbents used to enrich and separate various metal ions from sea water at trace levels. Uranium is one of the most valuable metals in seawater. Uranium is dissolved in seawater at a concentration of about 3 mg/m^3 in the ionic form of uranyl tricarbonate ion $UO_2(CO_3)_3^{4-}$ (Saito and Miyauchi, 1981, 1982). The selective recovery of uranium from seawater has been studied by many research groups particularly by the researchers from Takasaki Radiation Chemistry Research Establishment in Japan. The adsorbents they developed were mostly based on the synthesis of polymers bearing amidoxime groups, due to high affinity of this group in extraction of uranium from seawater (Saito and Sugo, 2000). Many types of polymeric adsorbents have been developed and tested for the recovery of uranium from sea water and aqueous media (Saraydın et al., 1995; Güven et al., 1999; Akkaş and Güven, 2000;

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Rivas et al., 2001). Most of these studies involve the incorporation of a pendant nitrile group into a polymer matrix, followed by the conversion of the nitrile group into amidoxime group by using the alkaline solution of hydroxylamine (Choi and Nho, 2000; Şahiner et al., 2000; Pekel et al., 2000; Riqueza et al., 2002). Considering the long term difficulties in the availability of uranium from conventional resources, most of these studies are centralized on the uranium adsorption from seawater by poly(amidoxime) polymer. Amidoxime chelate adsorbents typically contain functional amidoxime [$-C=NOH(NH_2)$] groups that can chelate with some metal ions, particularly with uranium and vanadium and create stable chelating systems, so that very pure metal ions can be obtained by elution (Egawa and Harada, 1979, 1980a, 1980b). Earlier work of Collega et al. showed that the adsorption of some transition, alkali and alkaline earth metal ions such as Na(I), K(I), Mg(II) and Ca(II) onto poly(acrylamidoxime) was negligible. It was found that amidoxime group exhibited no affinity for these metal ions (Collega et al., 1980). Similar results were also reported on the lack of affinity of amidoxime polymers in forming chelates with alkali and alkaline earth metal ions (Kantipuly et al., 1990; Garg et al., 1999). No attempt has therefore been made to investigate the adsorption of alkali and alkaline earth metal ions onto amidoximated poly(N,N′-dipropionitrile acrylamide) in this study.

Although a number of amidoximated adsorbents have been developed there are only few articles published for uranium uptake by poly(amidoxime) containing two amidoxime groups per repating unit (Kise and Sato, 1985; Park and Suh, 1996). Availability of two amidoxime groups per repeating unit is believed to make the adsorbent more accessible for the adsorption of metal ions in aqueous systems than the conventional adsorbents having only one amidoxime group per repeating unit. In our previous work, amidoximated poly(N,N′-dipropionitrile acrylamide) was synthesized as being the first acrylic bidentate amidoxime polymer in the amidoxime polymer family (Kavaklı Akkaş et al., 2004a). In another work, we prepared a new type of fibrous adsorbent with adjacent amidoxime groups by radiation induced graft polymerization (Kavaklı Akkaş et al., 2004b). The unique advantage of these polymers are that they contain double amidoxime groups per repeating unit, an additional diethylene spacer unit between neighbouring amidoxime groups in each monomeric unit to recover uranium and other transition metal ions from seawater and aqueous media at very low concentration levels more efficiently. By introducing more pendant amidoxime groups within the repeating unit of this new polymer, higher adsorption yields of uranyl and vanadyl ions have been anticipated.

In this work, the adsorption properties of amidoximated poly(N,N′-dipropionitrile acrylamide) in particulate form are investigated by a batch process for five different metal ions from very dilute aqueous solutions and also by using flow through mode for uranyl and vanadyl ions adsorption from seawater. The performance of this novel polymer was given herein.

Experimental

Materials

The novel chelating polymer used in this work was synthesized as follows: N,N′-dipropionitrile acrylamide (DPAAm) monomer was first synthesized by the reaction of 3,3-iminodipropionitrile and acryloyl chloride which were supplied by Aldrich Company and distilled under vacuum before use. The poly (DPAAm) was synthesized by free radical polymerization reaction using AIBN as the initiator and then synthesized polymer was reacted with hydroxylamine to obtain a polymer containing two amidoxime groups per repeating unit. Relevant experimental details have already been given elsewhere (Kavaklı Akkaş et al., 2004a).

1000 ppm standard solutions for chemical analyses from Cica-reagent Kanto Chemical-Co Inc. company were used for the adsorption studies of V(V), Cu(II), Co(II) and Ni(II) metal ions. In uranyl ion uptake studies standard solution containing 10 ppm of above mentioned metal ions were used by diluting to required concentrations.

Adsorption Experiments

The new polymer synthesized and amidoximated as described above was purified, dried and ground. Particles with size range of approximately 10 μm were used in adsorption experiments. A particular amount (around 0.015 g) of amidoximated poly(DPAAm) was treated with metal ion solutions (30 mL) in batch mode to get information on the relative performance of the amidoximated poly(DPAAm) adsorbent. The adsorption kinetics of uranyl, vanadyl, copper, cobalt and nickel ions from 100 ppb standard solutions were followed at pH 5 acetate buffer solution and 25°C. For this purpose, the amount of metal ions adsorbed was checked at 30 min, 1 h, 3 h, 6 h, 12 h and 24 h time intervals. After the adsorption equilibrium time was determined, the samples were contacted with five different concentration of metal ion solutions (3.3, 10, 100, 500 and 1000 ppb) at pH 5 acetate buffer with continuous stirring at 25°C for those predetermined time periods. Metal ion concentrations remaining in the solution after every adsorption stage were determined by using Hewlett Packard 4500 series Inductively Coupled Plasma analyser (ICP).

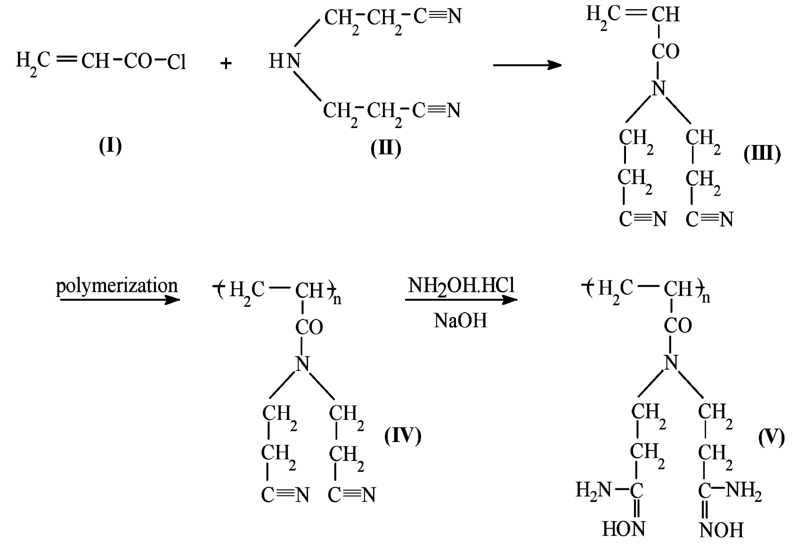
The selectivity for metal ion adsorption of amidoximated poly(DPAAm) was performed by using a mixture of aqueous solution of 100 ppb of each five metal ions following the procedure given above.

Seawater Treatment

The adsorption of uranyl and vanadyl ions from seawater was evaluated in flow-through mode. Approximately 0.05 g of amidoximated poly(DPAAm) particles was placed into a column with an inner diameter of 0.7 cm and height of 5 cm. Seawater sampled at the seashore of Mutsu Sekine-Hama in Aomori Prefecture was used after filtering through a 5 mm pore size paper filter (Tokyo Filter Co., Tokyo, Japan). Seawater was circulated upward through the amidoximated poly(DPAAm) loaded column at a flow rate of 6 mL/min at 25°C. After 1 hour, 1 day and 1 week contact time, the adsorbent was taken out from the column and immersed in 1 M HCl to elute the adsorbed uranyl and vanadyl. Amounts of adsorbed uranyl and vanadyl ions from eluted solutions were determined by ICP.

Results and Discussion

The synthesis of an acrylic polymer containing two adjacent amidoxime groups per repeating unit essentially involves three steps. The synthesis of a novel monomer, N,N′-dipropionitrile acrylamide, its conversion into polymer and further modification into amidoxime structures are shown in Scheme 1. The first step is the synthesis of monomer by an organic substitution reaction. The reaction of acryloyl chloride with 3,3-iminodipropionitrile results in the formation of N,N′-dipropionitrile acrylamide, an acrylic monomer with two pendant cyano groups. The second step is the polymerization of N,N′-dipropionitrile acrylamide by free radical mechanism using a thermal initiator. The polymer thus obtained with a maximum conversion of 77% is a white powder and soluble in several polar solvents such as dimethylformamide (DMF), dimethylsulfoxide (DMSO), tetrahydrofurane (THF), acetone, ethanol and chloroform. The final step is the amidoximation of cyano groups of poly(N,N′-dipropionitrile acrylamide). The cyano groups were converted to amidoxime groups (99%) by reacting with hydroxylamine hydrochloride in methanol solution, details related to



Scheme 1. Reaction scheme showing the synthesis of amidoximated poly(N,N′-dipropionitrile acrylamide).

the synthesis and characterization of this novel polymer have been given elsewhere (Kavaklı Akkaş et al., 2004a).

Batch Studies

In order to investigate the adsorption kinetics of five different metal ions (U(VI), V(V), Cu(II), Co(II) and Ni(II)) onto amidoximated poly(DPAAm), approximately 0.015 g of adsorbent was treated with 100 ppb metal ion solutions at pH 5 buffer solution at 25°C. Figure 1 shows the adsorption kinetics of U(VI), V(V), Cu(II), Co(II) and Ni(II) ions onto amidoximated poly (DPAAm). The adsorbed amounts are given as the percentage of metal ions initially present.

High adsorption rates are observed within 30 min for vanadyl ion and 1 h for uranyl ion and the plateau values (i.e., adsorption equilibrium) were reached approximately at 98% (210 μg/g) removal of vanadyl ion within 1 h and 98% (200 μg/g) removal of uranyl ion within 3 hrs.

The adsorption equilibria of Cu(II) and Ni(II) ions have been attained in about 30 min with 40% uptake (105 μg/g) for Cu(II) and in 3 hrs with 15.5% uptake (31.06 μg/g) for Ni(II) ions. The adsorption rate of Co(II) ion gradually increased and not reaching equilibrium even in 24 hrs. The adsorption value of 61% (123 μg/g) was reached for cobalt ions at the end of 24 hrs.

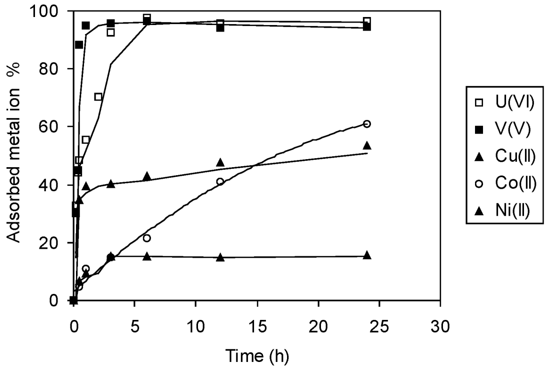


Figure 1. Adsorption kinetics of various metal ions from the same (100 ppb) initial concentrations.

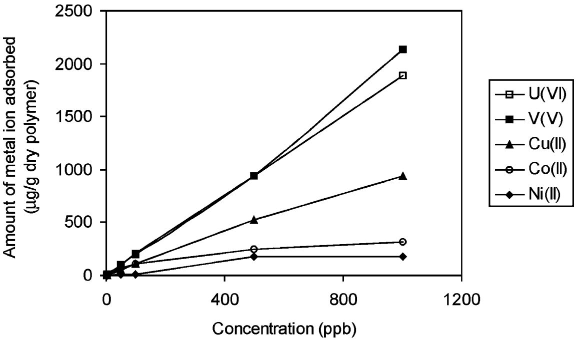


Figure 2. Dependence of metal ion uptake on the initial concentrations of metal ions.

The effect of initial concentration of metal ions on the adsorption behavior of amidoximated poly(DPAAm) (0.015 g) was determined in 6 hr contact times for U(VI), V(V), Cu(II), Ni(II) and 24 hr for Co(II) ion for five different metal ion concentrations (3.3, 50, 100, 500 and 1000 ppb) and given in Fig. 2. Figure 2 shows that adsorption of uranyl, vanadyl and copper ions increased almost linearly with increasing initial metal ion concentration. Adsorption of cobalt and nickel ions reached equilibrium after 500 ppb initial concentration.

The new adsorbent exhibited higher affinity for U(VI) and V(V) ions, the highest values were found to be 1887 μg/g (93%) and 2132 (98%) μg/g from 1000 ppb metal ions solution, respectively. Almost complete removal (100% adsorption) of vanadyl and uranyl ions have been reached easily for all concentrations studied. The amounts adsorbed for other metal ions from 1000 ppb initial concentration are 941 μg/g, 318 μg/g and 178 μg/g for Cu(II), Co(II) and Ni(II) ions corresponding to 64%, 16%, 10% adsorption of initial amounts, respectively.

The adsorption efficiency of newly synthesized adsorbent (double amidoxime functionality) was tested against the conventional one (mono amidoxime functionality) under identical adsorption conditions. For a range of initial uranyl ion concentrations (200–800 ppm) the average amount of uranyl adsorbed by the new adsorbent 322 mg/g, was found to be more than 50% higher than the corresponding value for conventional adsorbent, 212 mg/g dry adsorbent.

The adsorption capacity for uranyl ion by this new polymeric adsorbent containing two amidoxime groups per repeating unit is better than that of the chelating resins prepared by the method of Park et al. and Kise et al. (Park and Suh, 1996; Kise and Sato, 1985) which also contain enriched amidoxime groups. The unique advantage of the new polymer is that it contains double amidoxime groups per repeating unit, an additional diethylene spacer unit between neighbouring amidoxime groups in each monomeric unit. This polymer is known to be the first acrylic bidentate amidoxime polymer in the amidoxime polymer family reported in the literature. These properties make this new adsorbent more accessible for the adsorption of metal ions in aqueous systems than the conventional adsorbents having only one amidoxime group per repeating unit attached in close proximity to the backbone polymer.

Moreover applying the data of uranyl ion adsorption kinetics to different equations, adsorption isotherms were constructed for amidoximated poly(DPAAm) in a parallel study. Uranyl ion adsorption isotherms were found to agree very well with Langmuir type isotherm (Kavakli Akkas and Güven, 2004c).

Competitive Adsorption

The selectivity of amidoximated poly(DPAAm) is checked for 100 ppb mixture solution of U(VI), V(V), Cu(II), Co(II) and Ni(II) metal ions. Figure 3 shows the result of the selectivity experiment for amidoximated poly (DPAAm). The adsorption selectivity for a dilute solution of metal ions is expressed in the form of the distribution coefficient (D) which is a useful measure of affinity of an adsorbent (Sahni and Reedijk, 1984).

$$D = (\text{mg } M^{\text{n}+}/g \text{ of dry polymer})/ \\ (\text{mg } M^{\text{n}+}/\text{mL of solution})$$

The distribution coefficients D of each metal ion in the mixture solution under competitive conditions were determined by using the above equation. The results given in Fig. 3 are obtained at the end of 24 hrs of adsorption period. It can be seen from this figure that the new adsorbent selectively adsorbs uranyl ion compared to the other metal ions. The distribution coefficient of uranyl ion is 3 times higher than that for vanadyl ion and approximately 150 times higher than those of Cu and Co ions. There was a very big difference between nickel and other metal ions. Nickel ion was not adsorbed effectively. The selectivity order was found to be $U(VI) > V(V) \gg Co(II) = Cu(II) \gg Ni(II)$ for 100 ppb mixture metal ion solution. This can be explained by the following reasons; the amidoximes exist predominantly in the syn-hydroxyamino form which is stabilized by an intramolecular hydrogen bond (Rivas et al., 2000). They generally undergo a metal—assisted hydroxyl proton dissociation upon coordination and a rule of thumb is that 'soft acid' tends to interact with 'soft bases' and conversely, 'hard acids' interact with 'hard bases'. A classification of selected ions and ligands as hard and soft acid and bases (HSAB) according to the scale of Pearson was given in the literature (Pearson, 1963). The functional groups in the chelating polymer materials usually act as bases; oxygen and $-NH_2$ containing functional groups being hard. Uranyl and vanadyl ions act as hard acids, chelating polymers containing excess amidoxime groups having oxygen and $-NH_2$ containing functional groups synthesized in this work interact selectively and efficiently with uranyl and vanadyl ions. Ni(II), Cu(II) and Co(II) act as intermediate acids, $-NH_2$ and oxygen containing functional groups act as hard bases, consequently U(VI) and V(V) ions show strong interaction with both oxygen and $-NH_2$ containing functional groups than the other metal ions which have low interaction with oxygen and $-NH_2$ groups. A recent publication (Pletner and Zernov, 2002) on classification of metal ions according to their complexing properties prepared by evaluating stability constants of 24 metal ions with 3960 ligands classifies uranyl and vanadyl ions under the same category and Co(II), Cu(II), Ni(II) and Zn(II) under a different category.

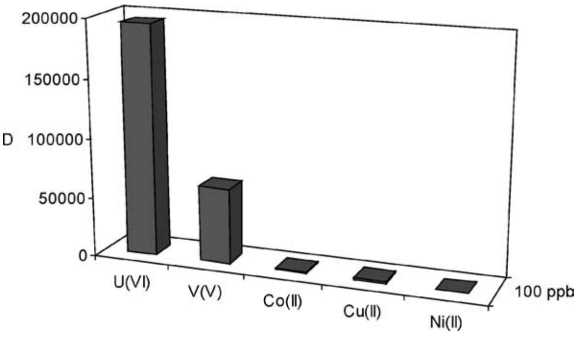


Figure 3. Adsorption selectivity expressed in terms of distribution coefficient D, of amidoximated poly(DPAAm) for the indicated metal ions.

Seawater Treatment

Uranyl and vanadyl ions adsorption from seawater were also evaluated in flow-through mode. After 1 hr, 1 day and 1 week contact time, adsorption capacity of amidoximated poly(DPAAm) were found to be 13.08, 28.1, 27.13 mg U/g of adsorbent and 30.36, 31.16, 33.27 mg V/g of adsorbent, respectively. These results shows that, after 1 day of contact time no appreciable increase in adsorption of V was observed and equilibrium

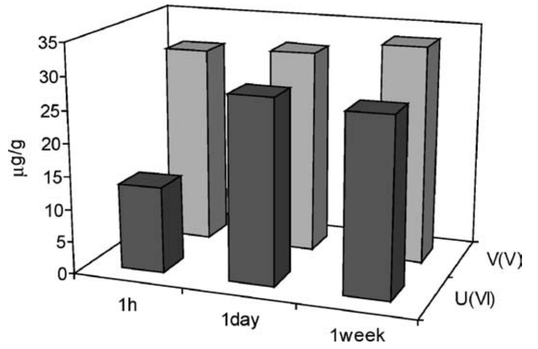


Figure 4. Adsorption of uranyl and vanadyl ion from seawater at different contact times.

degree of adsorption values were already reached. The same figure shows that amidoximated poly(DPAAm) has almost reached the adsorption equilibrium in 1 h for vanadyl and it was also observed that 3 hr was sufficient for maximum removal of uranyl ions. In terms of kinetic evaluation (rate of adsorption), batchwise adsorption of U(VI) and V(V) onto amidoximated poly (DPAAm) were nearly the same as with the sea water treatment process (Fig. 4). This new polymer has a higher vanadyl and uranyl ion adsorption capacity and adsorption rate from seawater than those that were developed by other groups.

Conclusions

A new polymeric adsorbent containing two amidoxime groups per repeating monomeric units has been shown to provide a rapid uptake within 1 h for the metal ions, U(VI), V(V), Cu(II), Co(II) an Ni(II) studied here and with high capacity for U and V ions. Approximately 100% adsorption of uranyl and vanadyl ions for all concentrations (3.3–1000 ppb) have been reached. From the competitive adsorption studies it was seen that the new adsorbent was quite selective for uranyl than the other metal ions with regard to corresponding distribution coefficients (D). The selectivity order determined by calculating D was found to be $\mathrm{U(VI)} > \mathrm{V(V)} \gg \mathrm{Co(II)} = \mathrm{Cu(II)} \gg \mathrm{Ni(II)}$. This new polymer has a higher vanadyl and uranyl ion adsorption capacity and adsorption rate from seawater. Finally, it can be said that this novel adsorbent containing two amidoxime group per repeating monomeric units is promising for the enrichment and separation of uranyl and vanadyl ion from seawater and aqueous solutions.

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