

# Removal of Concentrated Heavy Metal Ions from Aqueous Solutions Using Polymers with Enriched Amidoxime Groups

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Received 24 October 2003; accepted 2 March 2004

DOI 10.1002/app.20616

Published online in Wiley InterScience (www.interscience.wiley.com).

**ABSTRACT:** Particulate and fibrous adsorbents with enriched amidoxime groups were synthesized by using a novel monomer *N,N'*-dipropionitrile acrylamide. The adsorption properties of amidoximated poly(*N,N'*-dipropionitrile acrylamide) [poly(DPAAM)] particles and a nonwoven fabric grafted with the same for  $\text{UO}_2^{2+}$ ,  $\text{Pb}^{2+}$ ,  $\text{Cu}^{2+}$ , and  $\text{Co}^{2+}$  at high concentrations were investigated by batch process. Metal ion adsorption studies were conducted from metal ion solutions with different initial concentrations (100–1500 ppm). It was shown that particulated amidoximated poly-(DPAAM) has higher adsorption capacity than amidoxi-

mated nonwoven fabrics for all metal ions, especially for uranyl ions. The results of the adsorption studies showed that the interaction between  $\text{UO}_2^{2+}$  and amidoximated groups agree with the Langmuir-type isotherm. From the Langmuir equation, the adsorption capacities were found as 400 mg  $\text{UO}_2^{2+}$ /g dry amidoximated poly(DPAAM) and 250 mg  $\text{UO}_2^{2+}$ /g dry amidoximated graft polymer. © 2004 Wiley Periodicals, Inc. *J Appl Polym Sci* 93: 1705–1710, 2004

**Key words:** adsorption; fibers; functionalization of polymers; metal-polymer complexes

## INTRODUCTION

Metal ions are not only valuable intermediates in metal extraction, but also important raw materials for technical applications. Accordingly, complexation of metal ions is an important technique for recovering metals from various sources (hydrometallurgy) and for their removal from streams in municipal and industrial waste. As a consequence, complexation, separation, and removal of metal ions have become increasingly attractive areas of research and have led to new technological developments. Metal-chelating and ion exchange polymers were used in hydrometallurgical applications such as recovery of rare metal ions from seawater and removal of traces of radioactive metal ions from wastes.<sup>1</sup> These methods have found widespread applicability owing to their selectivity. A polymeric ligand is used to selectively bind a specific metal ion in a mixture to isolate important metal ions from wastewater and aqueous media.<sup>2–5</sup> A polymeric ligand is usually used in an insoluble resin form to separate a specific metal ion from a liquid containing a mixture of metal ions. For example, uranium is a

potential environmental pollutant, especially in mining industry wastewater, and the migration of uranium in nature is important in this context. In view of the anticipated exhaustion of terrestrial uranium reserve in the near future, research has been directed toward the recovery of uranium from nonconventional sources, such as coal and natural waters (0.1–10 mg U/m<sup>3</sup>), and especially from seawater (2.8–3.3 mg U/m<sup>3</sup>). The recovery of uranium from contaminated water of flooded mines (0.1–15 mg U/m<sup>3</sup>) also presents a very important environmental problem to be solved. Many types of adsorbents were developed and studied for the recovery of uranium from seawater and aqueous media.<sup>6–9</sup> Among them, amidoxime group containing adsorbents were shown to be the most effective for the recovery of uranium from seawater and aqueous media.<sup>10–17</sup> Egawa et al. in their pioneering work prepared a macroreticular chelating resin containing amidoxime by reacting acrylonitrile-divinylbenzene copolymer beads with hydroxylamine.<sup>18–20</sup> Amidoxime chelate fabrics were shown by Sugo and Saito and Lin et al. to be very useful for recovering uranium from seawater and enrichment or separation of noble metal ions from aqueous systems.<sup>21,22</sup>

Although a variety of amidoximated adsorbents were developed, there are very few studies published for uranium uptake by polyamidoxime resins containing two amidoxime group per repeating unit.<sup>23–24</sup> In our previous work, amidoximated poly(*N,N'*-dipropi-

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Contract grant sponsor: IAEA; contract grant number: TUR8016.

Contract grant sponsor: Academy of Sciences of Turkey.

onitrile acrylamide), which was the first acrylic bidentate amidoxime polymer in the amidoxime polymer family, was synthesized.<sup>25</sup> In another work, we prepared a new type of fibrous adsorbent with adjacent amidoxime groups by radiation-induced graft polymerization.<sup>26</sup> The unique advantage of these polymers is that they contain double amidoxime groups per repeating unit, an additional diethylene spacer unit between neighboring amidoxime groups in each monomeric unit to recover uranium and other transition metal ions from seawater, and aqueous media at very low concentration levels (ppb) more efficiently.<sup>27-28</sup>

In this study, adsorption capacity of newly developed adsorbent containing double amidoxime groups per repeating unit was investigated for uranyl, lead, copper, and cobalt ions from relatively concentrated (ppm) aqueous solutions under noncompetitive conditions. The higher concentration range was used to evaluate the performance of new adsorbent for the removal of relevant ions from relatively concentrated industrial and mining waste and process waters. The binding mechanism of metal ions with amidoxime groups were investigated by using FTIR. Moreover, applying the data of uranyl ion adsorption kinetics to different equations, adsorption isotherms were constructed for amidoximated poly(*N,N'*-dipropionitrile acrylamide) [poly(DPAAm)] and amidoximated nonwoven fabric.

## EXPERIMENTAL

### Materials

A novel adsorbent containing double amidoxime groups per repeating unit was synthesized with the anticipation of enhanced metal ion uptake capacity. For this purpose, *N,N'*-dipropionitrile acrylamide (DPAAm) monomer was first synthesized by the reaction of 3,3-iminodipropionitrile (Aldrich, Milwaukee, WI) and acryloyl chloride (Aldrich).<sup>25</sup> The poly(DPAAm) was synthesized from this monomer by free-radical polymerization by using AIBN (Merck, Darmstadt, Germany) as the initiator at 70°C with a conversion efficiency of 77%. The polymer synthesized was reacted with hydroxylamine (Aldrich) to convert the nitrile groups into amidoxime groups and 99% conversion was reached. The polymer was ground and microparticles were later used in adsorption experiments.

The preparation of nonwoven fabric grafted with side chains containing two amidoxime groups per monomeric unit required three steps. Glycidyl methacrylate (GMA) (Tokyo Kasei Co., Japan) was first radiation-grafted on polyethylene-coated polypropylene nonwoven fabric (Kurashiki Sen-I Kako Co., Okayama, Japan) and chemically modified with 3,3'-iminodipropionitrile [NH(—CH<sub>2</sub>—CH<sub>2</sub>—

CN)<sub>2</sub>] (IDPN) (Kanto Chemical, Tokyo, Japan). Grafted fabric was reacted with hydroxylamine to convert nitrile groups into amidoxime, consequently obtaining a new fibrous adsorbent containing two adjacent amidoxime groups in the repeating units of grafts. An amidoxime density of 2 mmol/g of dry fabric was achieved as reported in detail elsewhere.<sup>26</sup>

Metal salts used in the adsorption experiments Cu(NO<sub>3</sub>)<sub>2</sub>·3H<sub>2</sub>O, Co(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O were supplied by Merck and Pb(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O and UO<sub>2</sub>-(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O were supplied by BDH (Poole, UK).

Adsorption capacities of amidoximated poly(DPAAm) in powder form and grafted onto nonwoven fabric were determined for four different metal ions (UO<sub>2</sub><sup>2+</sup>, Cu<sup>2+</sup>, Pb<sup>2+</sup>, Co<sup>2+</sup>) from different initial concentrations (100–1500 ppm) by a batch process. Approximately 0.05 g dry amidoximated poly(DPAAm) was placed into 40 mL metal ion (UO<sub>2</sub><sup>2+</sup>, Cu<sup>2+</sup>, Pb<sup>2+</sup>, Co<sup>2+</sup>) solution buffered at pH 5 in a vial agitated magnetically at moderate rpm for a sufficient time. To determine the amount of metal ions adsorbed onto amidoximated poly(DPAAm), Co<sup>2+</sup>, UO<sub>2</sub><sup>2+</sup>, and Cu<sup>2+</sup> ion concentrations remaining in the solutions were determined by using Philips 8710 UV-Vis spectrophotometer by using sodium salicylate and ammonia as complexing agents for UO<sub>2</sub><sup>2+</sup> and Cu<sup>2+</sup> ions, respectively. The concentration of Pb<sup>2+</sup> ions was determined with an Hitachi AAS. Adsorption studies as mentioned above were also carried out for amidoximated nonwoven fiber (0.015 g).

The amounts of metal ion adsorbed per unit mass of the amidoximated poly(DPAAm) and nonwoven fiber were evaluated by using the expression

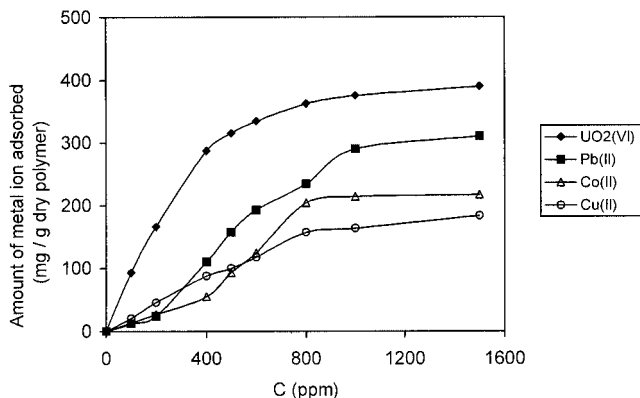
$$q_e = \frac{(C_0 - C_e)V}{W}$$

where  $q_e$  is the amount of metal ions adsorbed onto unit dry mass of amidoximated poly(DPAAm) or nonwoven fiber (mg/g),  $C_0$  and  $C_e$  are the concentrations of metal ion solutions initially and after treatment for a certain period of time (mg/L), respectively,  $V$  is the volume of aqueous phase (L), and  $W$  is the amount of dry amidoximated poly(DPAAm) or graft polymer used (g).

To investigate the interaction between metal ion and amidoximated polymers, FTIR spectra were taken on KBr discs by using a Nicolet 520 FTIR spectrophotometer.

## RESULTS AND DISCUSSION

Detailed information on the synthesis and amidoximation reaction of poly(DPAAm) and nonwoven fabric was given in our earlier publications.<sup>25,26</sup> Certain amounts of amidoximated poly(DPAAm) powder and



**Figure 1** Dependence of metal ion uptake on the initial concentrations of metal ions for amidoximated poly(DPAAM).

amidoximated nonwoven fabric were put into different concentrations (100–1500 ppm) of four different metal ion solutions at pH 5 under continuous agitation until they reached adsorption equilibrium. The binding properties of these metal ions (i.e., UO<sub>2</sub><sup>2+</sup>, Pb<sup>2+</sup>, Cu<sup>2+</sup>, and Co<sup>2+</sup>) with amidoximated poly(DPAAM) powder and amidoximated nonwoven fabric determined in different concentrations of aqueous metal solutions at pH 5 acetate buffer solution are given in Figures 1 and 2.

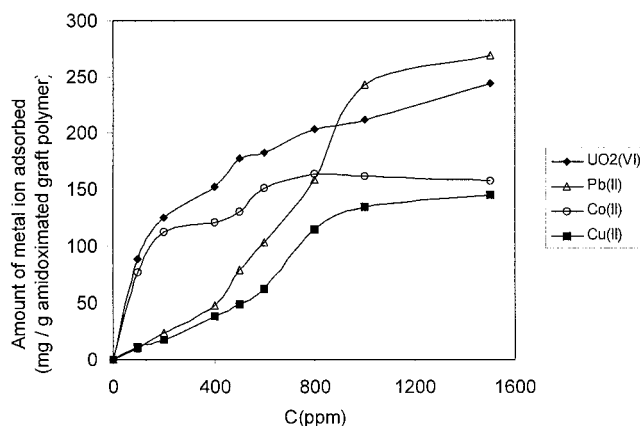
It was found that the metal ion uptake by amidoximated poly(DPAAM) powder increased with increasing initial metal ion concentrations and beyond 1000 ppm metal ion concentration adsorption equilibria were reached, as shown in Figure 1. At this point, the amount of adsorbed uranyl ion was determined to be 390 mg UO<sub>2</sub><sup>2+</sup>/g dry amidoximated polymer. The adsorption behavior of Pb<sup>2+</sup>, Cu<sup>2+</sup>, and Co<sup>2+</sup> ions, which have lower adsorption capacities compared to uranyl, shows similar adsorption isotherms with UO<sub>2</sub><sup>2+</sup> ion. It was found that the adsorption capacity of amidoximated poly(DPAAM) for Pb<sup>2+</sup>, Co<sup>2+</sup>, and Cu<sup>2+</sup> ions for 1500 ppm initial concentration was 310, 217, and 184 mg metal ion/g dry polymer, respectively. The order of adsorption affinity based on the amount of metal ion uptake (mg metal ion/g dry polymer) is as follows: UO<sub>2</sub><sup>2+</sup> > Pb<sup>2+</sup> >> Co<sup>2+</sup> > Cu<sup>2+</sup>.

UO<sub>2</sub><sup>2+</sup>, Pb<sup>2+</sup>, Cu<sup>2+</sup>, and Co<sup>2+</sup> ion adsorption capacities of the amidoximated nonwoven fabric as a function of initial concentration of metal ions were given in Figure 2. As shown in this figure, amidoximated nonwoven fabric showed high affinity for Pb<sup>2+</sup> ion as well as UO<sub>2</sub><sup>2+</sup> ion. Cobalt and copper ions have lower adsorption capacities than lead and uranyl ions. The amount of metal ions adsorbed per unit mass of the polymer (i.e., adsorption capacity) increased first with increasing the initial concentration of metal ions and then reach the plateau value, which represents satu-

ration of the active points (which are available for specific metal ions) on the polymers. It was found that the adsorption capacity of amidoximated poly(DPAAM) for Pb<sup>2+</sup>, UO<sub>2</sub><sup>2+</sup>, Cu<sup>2+</sup>, and Co<sup>2+</sup> ions at 1500 ppm was 267, 243, 145, and 157 mg metal ion/g dry amidoximated graft polymer, respectively. The order of affinity based on the amount of metal ion uptake (mg metal ion/g dry polymer) is as follows: Pb<sup>2+</sup> > UO<sub>2</sub><sup>2+</sup> >> Co<sup>2+</sup> > Cu<sup>2+</sup>.

The amidoximated poly(DPAAM) has higher adsorption capacity for all metal ions studied in this work than the amidoximated nonwoven fabric, as shown in Figures 1 and 2. The explanation can be given as follows: poly(DPAAM) was obtained as a homopolymer directly from *N,N'*-dipropionitrile acrylamide monomer but the amidoximated nonwoven fabric was first grafted with GMA and then functionalized with 3,3'-iminodipropionitrile, which was not 100% complete. The conversion of nitrile groups into amidoxime for poly(DPAAM) was higher than for amidoximated nonwoven fabric.

An extensive sorption study on polyamidoxime polymer obtained from the copolymer of acrylonitrile-divinylbenzene was undertaken especially for extraction of uranyl ions from aqueous solutions by Egawa et al and Kabay et al.<sup>29,30</sup> They found that the sorption capacity of uranyl ion was about 310 mg/g (the initial concentration of UO<sub>2</sub><sup>2+</sup> ion solution was 2700 ppm). Vernon and Shah prepared a chelating polymer bearing hydroxamic/amidoxime groups and the binding of copper and uranium was observed to be 165 and 214 mg/g, respectively.<sup>31</sup> Lutfor et al. studied the adsorption of uranyl ions onto polyacrylonitrile-grafted sago starch and it was found that uranyl ion uptake by this polymer was 283 mg/g.<sup>32</sup> Rivas et al. used a commercial poly(amidoxime) chelating polymer and investigated the adsorption capacity of Cu, Cd, Hg, Pb, and U ions by batch technique.<sup>33</sup> It was



**Figure 2** Dependence of metal ion uptake on the initial concentrations of metal ions for amidoximated nonwoven fabric.

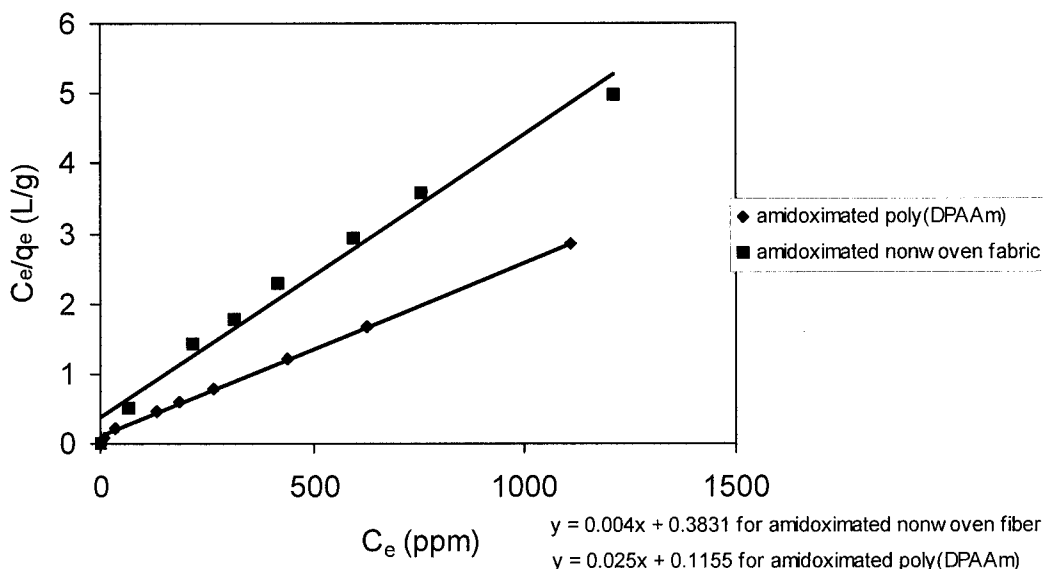


Figure 3 Langmuir isotherms for  $\text{UO}_2^{2+}$  ion adsorption on amidoximated poly(DPAAm) and nonwoven fabric.

found that this resin exhibited affinity for  $\text{UO}_2^{2+}$  and  $\text{Pb}^{2+}$ , but no affinity for the other metal ions. Adsorption capacity of this polymer from 4.98 mM (1344 ppm) solution of uranium was 130 mg/g. In this study, adsorption capacities of the newly synthesized adsorbents from 1500 ppm solution of uranyl ion was found to be 390 mg/g for amidoximated poly(DPAAm) and 243 mg/g for amidoximated graft polymer. In light of these findings, it can be said that bidentate amidoxime polymers have great advantage over the monodentate amidoxime polymer.

Adsorption isotherms of amidoximated poly(DPAAm) and amidoximated nonwoven fabric were analyzed according to the linear form of the Langmuir isotherms for uranyl ion, using the expression

$$\frac{C_e}{q_e} = (1/K_L q_{\text{mon}}) + (1/q_{\text{mon}})C_e$$

where  $C_e$  is the equilibrium concentration of adsorbate in solution,  $q_e$  represents the adsorbed  $\text{UO}_2^{2+}$  ion per unit mass of adsorbent at equilibrium,  $q_{\text{mon}}$  denotes the amount of adsorption corresponding to complete monolayer coverage, and  $K_L$  is the Langmuir constant. A plot of  $C_e/q_e$  versus  $C_e$  would give  $K_L$  and  $q_{\text{mon}}$ .<sup>34</sup>

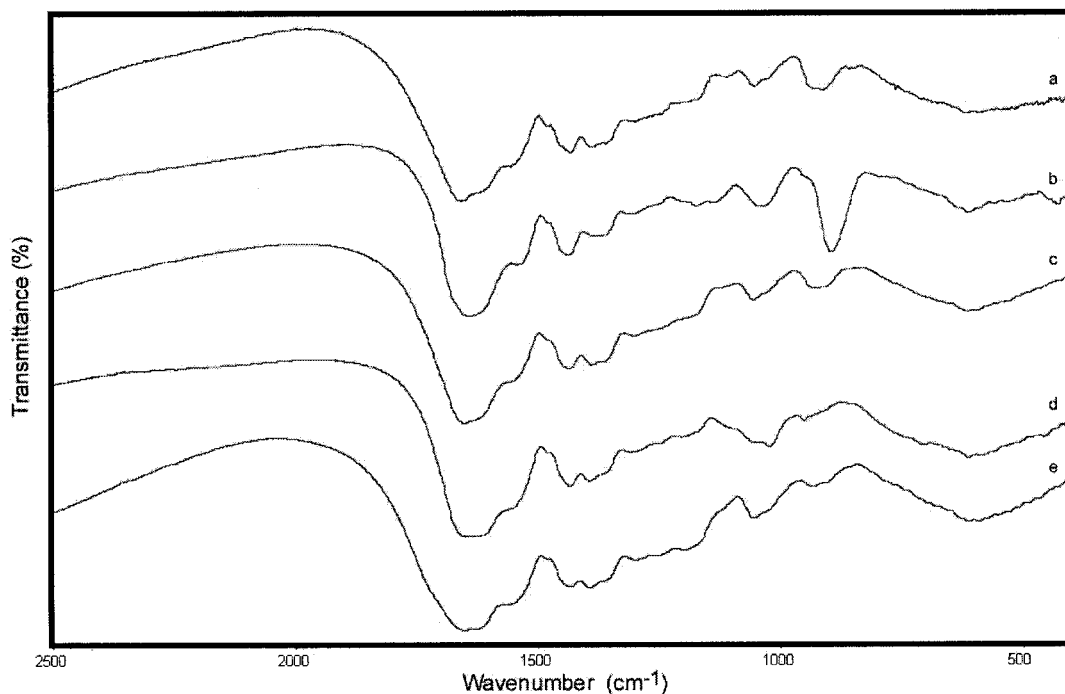
The plots of isotherms are shown in Figure 3 and seen to be linear over the whole concentration range studied.  $K_L$  and  $q_{\text{mon}}$  values calculated from Langmuir equation are given in Table I. The constant,  $K_L$ , contains enthalpic interaction of the binding of metal ions with polymer and can be found from the intercept of the lines given in Figure 3. As can be seen from Table I, the  $K_L$  value of amidoximated poly(DPAAm) powder is bigger than amidoximated nonwoven fabric, which indicates the favorable in-

teraction of accessible amidoxime sites on the surface of the polymer.  $q_{\text{mon}}$  is a quantity representing adsorption capacity, also known as monolayer coverage of the surface.  $q_{\text{mon}}$  values of amidoximated polymer and nonwoven fabric from the Langmuir equation for  $\text{UO}_2^{2+}$  ions were found to be 400 and 250 mg/g, respectively. This is in good accordance with the results previously found and discussed in this study.

To understand the possible interaction between metal ions and amidoxime groups of the adsorbents used in this work, FTIR spectra of samples before and after metal ion adsorption were taken and are shown in Figure 4. Due to interactions with  $\text{UO}_2^{2+}$  ions, there is a shift in the N—O characteristic band of polymers. As can be seen from Figure 4, after the adsorption of  $\text{UO}_2^{2+}$  ion, the N—O band initially observed at  $927\text{ cm}^{-1}$  shifts to  $899\text{ cm}^{-1}$ , as a result of interaction of corresponding groups with  $\text{UO}_2^{2+}$ . It has already been shown earlier that, as the metal ion—oxygen (M—O) bond becomes stronger, corresponding bands exhibit a lower stretching frequency.<sup>35</sup> These differences can be better seen in the difference spectrum given in Figure 5, especially with the appearance of a new peak

TABLE I  
 $K_L$  and  $q_{\text{mon}}$  Values Determined from Figure 3 for the Adsorption of  $\text{UO}_2^{2+}$

For $\text{UO}_2^{2+}$ adsorption	Amidoximated poly(DPAAm) powder	Amidoximated nonwoven fiber
$K_L$	$2.16 \times 10^{-2}$	$1.04 \times 10^{-1}$
$q_{\text{mon}}$	400	250

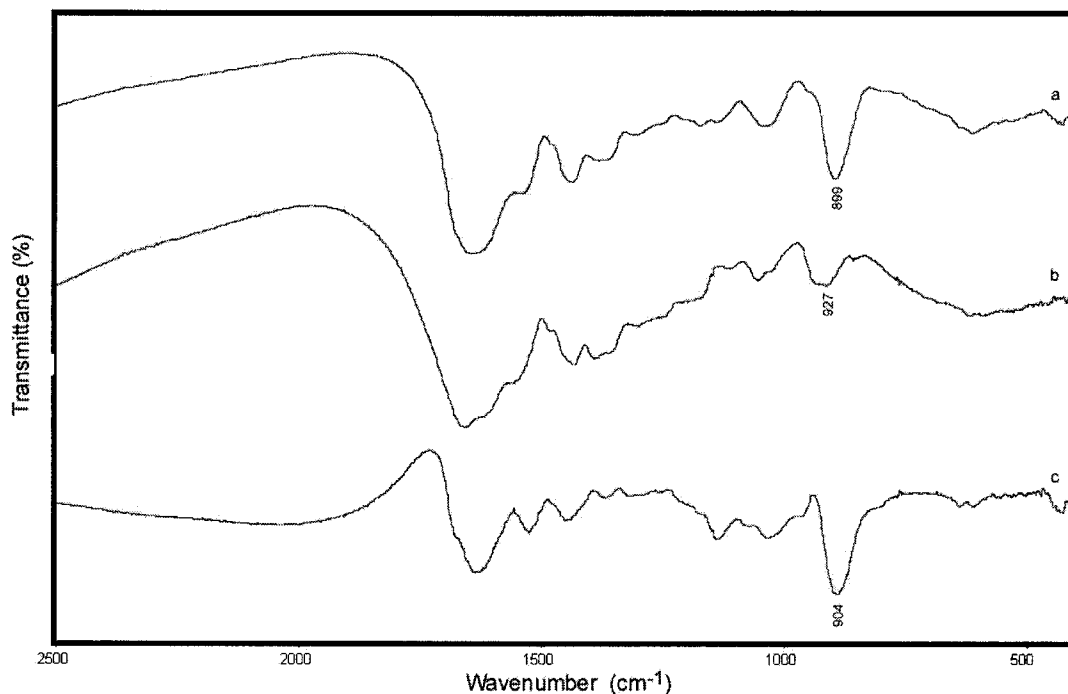


**Figure 4** FTIR spectra of (a) unadsorbed (b)  $\text{UO}_2^{2+}$  ion adsorbed, (c)  $\text{Pb}^{2+}$  ion adsorbed, (d)  $\text{Cu}^{2+}$  ion adsorbed, (e)  $\text{Co}^{2+}$  ion adsorbed amidoximated poly(DPAAm).

at  $904\text{ cm}^{-1}$  assigned to O—U—O stretching vibration.<sup>7,32</sup>

Similar differences were observed in the FTIR spectra of adsorbents containing other metal ions. For the

spectrum of adsorbed Pb, Cu, and Co sample, N—O band initially around  $927\text{ cm}^{-1}$  becomes weaker and shifts to around  $920\text{ cm}^{-1}$ . The FTIR spectra of amidoximated nonwoven fabric before and after metal



**Figure 5** FTIR spectra of (a)  $\text{UO}_2^{2+}$  ion adsorbed and (b) unadsorbed amidoximated poly(DPAAm), (c) spectral difference between (a) and (b).



ions adsorption is not given in the article because similar behaviors (shifts) were observed for uranyl ion and other metal ions. All the above results proved the interaction between the metal ions and amidoximated polymer.

### CONCLUSION

The adsorption capacity of amidoximated poly(D-PAAm) powder and amidoximated nonwoven fabric was investigated for  $\text{UO}_2^{2+}$ ,  $\text{Pb}^{2+}$ ,  $\text{Cu}^{2+}$ , and  $\text{Co}^{2+}$  metal ions at a high concentration level. The order of affinity based on amount of metal ion uptake (mg metal ion/g dry polymer) is as follows:  $\text{UO}_2^{2+} > \text{Pb}^{2+} \gg \text{Co}^{2+} > \text{Cu}^{2+}$  for amidoximated poly(DPAAm) powder and  $\text{Pb}^{2+} > \text{UO}_2^{2+} \gg \text{Co}^{2+} > \text{Cu}^{2+}$  for amidoximated nonwoven fabric at concentrations  $> 1000$  ppm metal ion.

The result of uranyl ion adsorption studies for two adsorbents complied with the L-type isotherm. From the Langmuir equation, the adsorption capacities were found to be 400 mg  $\text{UO}_2^{2+}$ /g dry polymer and 250 mg  $\text{UO}_2^{2+}$ /g dry polymer for amidoximated poly(D-PAAm) and nonwoven fabric, respectively.

It was concluded from these results that the adsorption capacities of these new poly(amidoxime) chelating polymers for uranyl ions are very high. These new adsorbents are more accessible for the adsorption of metal ions in aqueous systems than the conventional adsorbents having only one amidoxime group per repeating unit.

One of the authors (P.A.K.) thanks IAEA (Grant TUR8016) for the fellowship provided to carry out part of this work at the Takasaki Radiation Chemistry Research Establishment of JAERI. The authors are also grateful to Masao Tamada, Noriaki Seko, Haruyo Amada, and Fatmuanis Basuki for help with some of the experiments. Continuous interest and support by Takanobu SUGO are greatly appreciated. O.G. acknowledges the support of Academy of Sciences of Turkey.

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