A Study in the Uranyl Ions Uptake on Acrylic Acid and Acrylamide Copolymeric Hydrogels

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ABSTRACT: A series of copolymeric hydrogels comprising of acrylic acid and acrylamide and crosslinked with trimethylolpropane triacrylate (TMPTA) were prepared using ammonium persulfate (APS) as initiator. The hydrogels were functionalized further by partial hydrolysis and were characterized by SEM, FTIR, nitrogen analysis, and also by water uptake studies as a function of time, temperature, pH, NaCl, and concentration of sodium dodecyl sulfate (SDS). These hydrogels were used as sorbents for the uranyl ions uptake in the presence of 5% NaCl, which was studied as function of time, temperature, pH, and ion strength. The uranyl uptake was found to be affected both by the structural aspects of the hydrogels as well as by the external environmental factors. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 110: 3795–3803, 2008

Key words: hydrogels; polymer analogous reaction; sorption; TMPTA; uranyl ions

INTRODUCTION

Extraction of uranium from sea water presents a very important problem to be solved because of the growing demand of nuclear energy as clean source of energy. Further, actinides pose serious problem of toxicity and radioactivity. Sorption with polymeric materials is considered to be an effective method for the extraction of uranium because of the ease of handling, environmental safety, and high selectivity for uranium.[1,2] Polymeric and copolymeric networks with desirable functionalities are being continuously developed for the complexation with metal ions for sorption studies.[3–6] The copolymer of acrylamide (AAm) with the diprotic acid has been reported as adsorbent in the adsorption of some heavy metal ions.[7] Polyacrylonitrile fibers containing amidoxime group were used by Omichi et al.[8] for uranium recovery from sea water. Amidoxime derivatives of copolymeric hydrogels prepared from acrylonitrile and divinyl benzene have been used in the extraction uranium from sea water.[9] Sorption of uranium from poly(N-vinyl-2-pyrolidone-g-citric acid) hydrogels were reported by Caykara et al.[10] In recent years, prestructuring of hydrogels have been developed for the enhancement of uranyl ions uptake.

Ozyurek et al.[11] developed hydrogels by prestructuring of hydrogels to increase the capacity and selectivity of uranyl ions in the presence of lead and cadmium ions.

In view of the above, and in continuation to our earlier work on the use of hydrogels for the separation, enrichment and removal of metal ions from their aqueous solutions,[12–15] in this article we report preparation of the environment responsive hydrogels based on acrylic acid (AAc) and AAm, i.e., poly(AAc-co-AAm) and prepared by variation of the concentration of trifunctional crosslinker [trimethylolpropane triacrylate (TMPTA)]. To make these hydrogels more efficient to the uranyl uptake, further functionalization by polymer analogous reaction, i.e., partial alkaline hydrolysis was affected. The synthesized hydrogels were characterized by physical and chemical methods including scanning electron micrography (SEM), nitrogen analysis, and Fourier transformed infrared (FTIR) spectroscopy to establish their structure property relationship. These hydrogels were used for the sorption of uranyl ions in the presence of 5% NaCl, on analogy of the presence of the uranyl ions in sea water. The enrichment of uranyl ions from the sea water poses a technological challenge in view of the concentration of other competing ions. Further, the use of hydrogels under high electrolyte concentrations like sea water, results in the deswelling or low water interaction, hence poor partitioning of the metal ions. Hence, results of the uranyl ions uptake in brine solution as presented in this article are of interest.

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EXPERIMENTAL

Materials

Acrylamide (AAm), acrylic acid (AAc), ammonium persulfate, sodium dodecyl sulfate (SDS), sodium hydroxide, uranyl nitrate, sodium chloride, sodium carbonate, and buffer tablets all from S.D. Fine Chem., Mumbai, India and TMPTA from Sigma-Aldrich, New Delhi were used as received.

Synthesis of the networks

Equimolar amount of AAc and AAm were added to 10 mL of water in the presence of ammonium persulfate (APS) (1% by weight of both the monomers). To the reaction system was added TMPTA (1, 2, and 4% by weight of the monomers). The reaction mixture was stirred before it was pushed into glass ampoules under vacuum. The ampoules were sealed and were placed in a temperature controlled water bath at 50°C, and the reaction was allowed to proceed for 30 min. The networks thus prepared were cut into small pieces of equal dimensions, and were washed with water to remove any soluble fraction, dried at 40°C in air oven and then characterized. Partial hydrolysis of hydrogels was carried out by immersion of known weight of each network in 50 mL of 0.5M NaOH for 48 h. The networks were washed, dried as discussed above, and designated as poly(AAc-co-AAm)-cl-TMPTA.

Characterization of hydrogels

Characterization of the functionalized hydrogels was carried by SEM (Joel Stereoscan-150 microscope), FTIR spectra were taken on Nicollet 5700, and nitrogen analysis on Carlo Erba Instrument 1150. To study the swelling behavior of these hydrogels, dry gels were placed in double distilled water and swelling experiments were recorded at different conditions of time (10, 30, 60, 120, 240, 360, 600 min), temperature (25°C, 30°C, 35°C, 40°C, 45°C) with sensitivity of water bath ± 0.1°C, pH (4.0, 7.0, 9.0), and surfactant (SDS) concentrations (4.05, 8.32, 12.15 mM/L). The percent swelling of each hydrogel was calculated from the following relation:

$$P_s = ((M_t - M_0)/M_0) \times 100$$

where M_t is the mass of swollen gel at time t and M_0 is the initial mass of the dry gel. The salt sensitivity of the hydrogels was calculated as per the following relationship:

f(salt sensitivity)

$$= 1 - (\text{water uptake in a salt solution}$$
$$/\text{water uptake in distilled water})$$

Uranyl uptake studies

Uranyl ions uptake from aqueous solutions was investigated in continuous adsorption-equilibrium experiments. For this purpose, 0.15 g of dry hydrogels were transferred into a 50 mL of uranyl solutions in the concentration range 251–600 ppm of $UO_2(NO_3)_2$ in Na_2CO_3 buffer at pH 13.4 (0.1N Na_2CO_3) and 5% by weight of NaCl and allowed to equilibrate for different conditions of time and temperature while being shaken continuously. The aqueous solutions were separated from the hydrogels at definite time intervals and spectrophotometric measurements were carried out using a Cary 300 UV/Visible spectrophotometer at an absorption maximum of 238 nm. The amounts of adsorption per unit mass of the poly(AAc-co-AAm) hydrogels were evaluated by using the following expression:

$$q_e = (C_0 - C_e)V/W$$

where, q_e is the amount of uranyl ions adsorbed onto unit dry mass of the poly(AAc-co-AAm) hydrogels (mg g^{-1}), C_0 and C_e are the concentrations of uranyl ions in the initial solution and in the aqueous phase after treatment for a certain period of time, respectively, V is the volume of the aqueous phase and W is the amount of dry poly(AAc-co-AAm) hydrogels used (g).
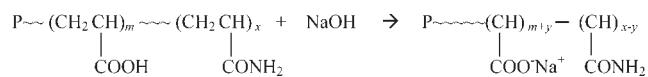
RESULTS AND DISCUSSION

The network formed by the use of a free radical initiator like APS is a clean and least energy consuming process as it dissociates at the ambient temperature in the presence of water. The presence of a trifunctional crosslinker like TMPTA provides the fast curing, heat stable, light resistant, solvent resistant, highly brittle performance properties to the gels because of sterically hindered backbone.[16]

The reactivity of monomers in copolymerization reaction even in the presence of crosslinker depends upon their reactivity ratio and so does the proportion of monomers in the macroradicals formed.[17] The reaction sites from the growing polymer result in the formation of crosslinked network with polymer and monomer and the resulting growing three-dimensional network can be represented as:
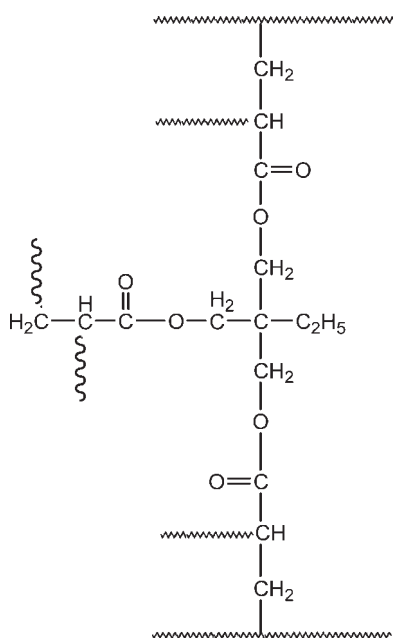
Partial hydrolysis of networks

Partial hydrolysis by way of saponification results in more active centers by charge generation, hence hydrogel-metal ions interactions get enhanced.[12] In the present study, partial hydrolysis affect both —CO_2H and amide groups and these get converted to carboxylate ions. The interactions of the ionic and ionogenic groups results in more water uptake by the hydrogel and also enable the hydrogel to partition metal ions to the polymeric phase. The generalized reaction scheme for the functionalization by partial hydrolysis where some of the amide groups get partially hydrolyzed to carboxylate groups is shown as:

$$P\sim(CH_2CH)_m \sim\sim (CH_2CH)_x + NaOH \rightarrow P\sim\sim\sim(CH)_{m+y} - (CH)_{x\text{-}y}$$

where the COOH and CONH_2 groups on the left and COO^-Na^+ and CONH_2 on the right are shown below the chain.

where m and x is the degree of polymerization of AAc and AAm, respectively, in the network and y is conversion of amide groups to carboxylate and $m + y$ is the total number of carboxylate groups present, assuming the whole of the poly(AAc) reacts with the alkali. Conversion of amide groups is always partial due to the electrostatic and steric factors under operating conditions.[18] Since ester groups are also liable to hydrolysis, hence some of these may get converted to carboxylate groups. However, the extent of such reaction is very low as most of these groups are not accessible to the alkali present in the reaction medium, especially at the crosslinks. The possibility of conversion of these groups should be minimal in the networks prepared with higher crosslinker concentration, as higher crosslinking density reduces the conversion rates due to the problem of inaccessibility to the reactive sites.[19]

Characterization of crosslinked polymers

SEM, FTIR spectroscopy, and nitrogen analysis have been used to characterize the networks. A comparison of the effect of the synthetic conditions on the structure of hydrogel has been studied.

Scanning electron micrography

Figure 1(a–d) reveals surface morphology of different polymeric networks. It provides evidence of pore formation as well as partial hydrolysis. Pore formation due to the crosslinking reactions is visible in the micrographs. The effect of crosslinker concentration on the surface morphology is also evident as pore size becomes smaller in the case of hydrogels prepared with the higher crosslinker concentrations. Considerable morphological changes are brought about after hydrolysis through these reactions in the polymer backbones, as the surface gets disrupted due to the hydrolysis, and also the pore size become bigger than those are noticeable in the precursor networks.

FTIR spectra analysis

The evidence of monomer and crosslinker incorporation onto polymer was obtained by analyzing the characteristic peaks of the functional groups presents in the spectra of the polymer synthesized. FTIR spectra of poly(AAc-co-AAm)-cl-TMPTA crosslinked networks are presented in Figure 2. All the crosslinked networks show prominent peaks in the region 3446–3445 cm^{-1} due to N—H stretching of amides, and there are prominent peaks at 1654.2 cm^{-1} (due to C=O stretching of amide), intense peaks around 1600 cm^{-1} and 1456.5–1455 cm^{-1} is ascribed due to C=O stretching of carboxylate ions, and characteristic band at 1741.3 cm^{-1} due to the stretching of C=O bond of ester, i.e., TMPTA. The partially hydrolyzed polymers show all the above mentioned peaks with weaker ester peak.

Nitrogen analysis

Results of nitrogen analysis of unhydrolyzed and partially hydrolyzed polymers are presented in Table I. The presence of nitrogen confirms formation of the network. Percent nitrogen remains same, irrespective of the percent of TMPTA, hence it is suggested that the monomers react as per their reactivity ratio. From the amount of nitrogen found, the weight ratio of AAm and AAc were calculated (Table I). The mole fractions of the two monomers have been calculated from the obtained nitrogen values. The ratios are close to the reported monomer reactivity ratio of these monomers.[20] From the calculated
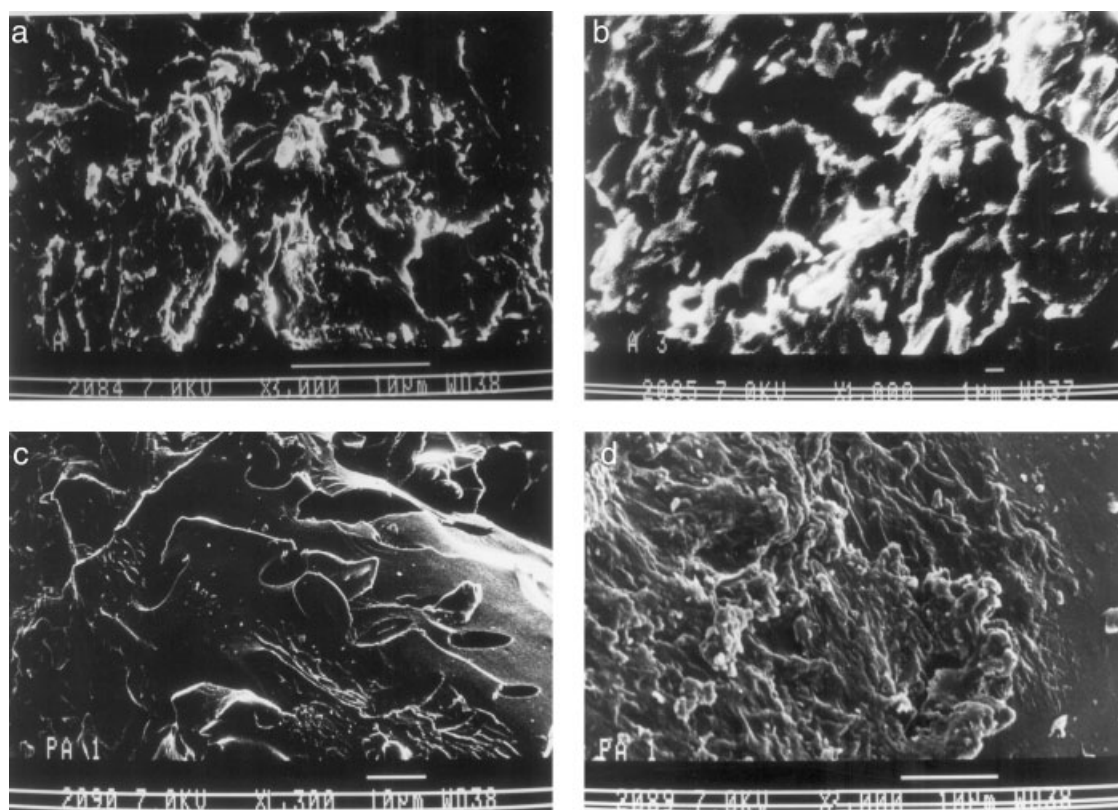
Figure 1 Scanning electron micrograph of poly(AAc-*co*-AAm) based hydrogels. (a) 1%, (b) 4% crosslinker TMPTA and partially hydrolyzed poly(AAc-*co*-AAm)-*cl*-TMPTA based hydrogels, (c,d) 1% crosslinker TMPTA at different magnifications.

mole fraction, it is observed that the reactivity ratio of AAc is greater than unity. Hence, the probability of AAc entering into the copolymer chain is higher as compared to AAm. Therefore, the copolymer formed is richer in AAc.

The loss of nitrogen is observed in all cases after hydrolysis. It confirms conversion of some amide groups to carboxylate groups. Poly(AAc-*co*-AAm)-*cl*-TMPTA prepared with 1% TMPTA has 8.35% nitrogen while corresponding hydrolyzed polymers has very low percent of nitrogen, i.e., 2.71% only, providing a strong evidence for partial hydrolysis. Similar trends are obtained with networks prepared in the presence of 2% and 4% crosslinker. From the loss of nitrogen on saponification, percent conversion of the amide groups of poly(AAm) to carboxylate groups has been calculated and presented in (Table I). The data shows that under the conditions employed, hydrolysis was partial.

Swelling behavior of poly(AAc-*co*-AAm)-*cl*-TMPTA hydrogels

Study of the water uptake behavior of the hydrogels is important to design hydrogels for metal ion uptake. It is evident from Figure 3(a–h) that both structural as well as environmental factors strongly
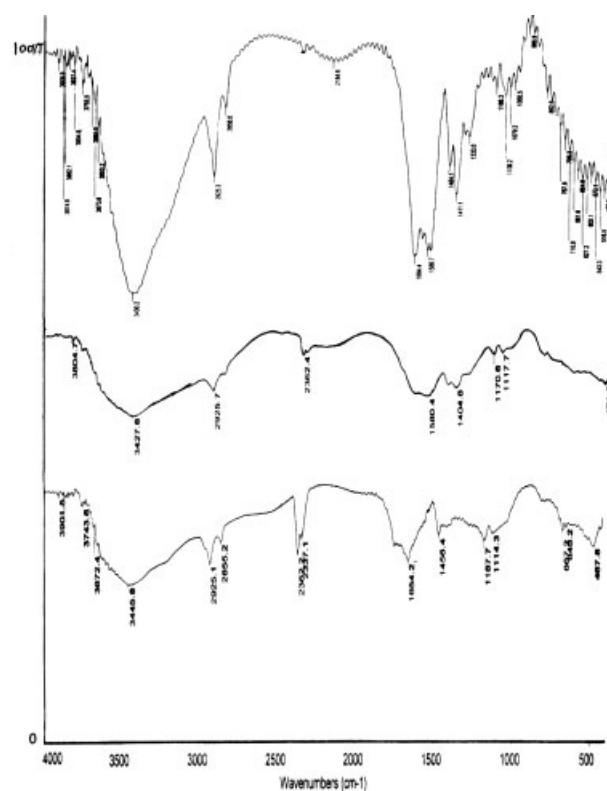
F3

Figure 2 FTIR of poly(AAc-*co*-AAm)-*cl*-TMPTA: (1) unhydrolyzed, (2) hydrolyzed, and (3) uranyl loaded hydrogels.

TABLE I
Elemental Analysis of Hydrogels

Crosslinker concentration (%)	Wt. of sample (mg)	Nitrogen found (%)	Wt. of nitrogen (mg)	Wt. of AAm (mg)	Weight ratio (AAm : AAc)	% Hydrolysis	Mole fraction of unhydrolyzed AAm
Poly(AAc-co-AAm)-cl-TMPTA							
1	2.514	8.35	0.1799	0.9124	1 : 1.70	–	0.38
2	1.989	8.10	0.1611	0.8170	1 : 1.36	–	0.43
4	2.347	8.32	0.1953	0.9901	1 : 1.25	–	0.45
Partially hydrolyzed poly(AAc-co-AAm)-cl-TMPTA							
1	2.355	2.71	0.0638	0.3236	1 : 6.13	63.16	0.14
2	2.167	2.87	0.0622	0.3154	1 : 5.67	65.12	0.15
4	2.166	3.09	0.0669	0.3393	1 : 5.07	62.22	0.17

influence the swelling kinetics and percent swelling of different hydrogels. Swelling behavior of poly(-AAc-co-AAm)-cl-TMPTA has been studied as a function of crosslinker concentration, effect of hydrolysis, and also of the external environmental factors like pH, temperature, and nature of the swelling medium. The swellability of poly(AAc-co-AAm)-cl-TMPTA decreases with the increase in crosslinker concentration [Fig. 3(a–h)]. This observation shows that the crosslinking density increases with the increase of crosslinker concentration, hence, with that the pore size of the crosslinked network decreases. Similar conclusion has been drawn by Chancy and Pillai[21] and Naghash and Okay.[22]

The effect of the time on the swelling behavior of different networks is presented in Figure 3(a,b). Swelling was studied at eight different time intervals from 10 to 600 min, at 25°C. Generally, all the crosslinked networks show initial increase in P_s with increase in time. Thereafter, swelling becomes almost constant after attaining equilibrium swelling. Swelling behavior of different crosslinked polymers was also studied at five different temperatures, i.e., 25–45°C with the successive increment of 5°C at different time intervals. All the networks show almost same trends as described above for the swelling behavior at 25°C, while the maximum P_s was observed at 40°C. In most of the hydrogels the rate of swelling changes abruptly on variation of time and temperature. The equilibrium swelling is attained at the higher swelling time and temperature. From the foregone discussion, it is clear that at 360 min and 40°C all the hydrogels show maximum swelling.

The effect of variation of pH on P_s was studied in the range 4.0–9.0 at 40°C. P_s increases on moving from acidic to neutral medium, and then, decreases from neutral to basic medium [Fig. 3(c)]. The behavior is almost common for all the hydrogels irrespective of the concentration of the crosslinker. These trends in swelling behavior originate from the suppression of the ionization of the carboxylic groups at low pH where shriveling of the hydrogels was observed. In alkaline pH, the hydrogels interact with

the basic species again restricting the interaction of water molecules with the functional groups of network that may result from preference of hydrogel for the basic moiety of buffer.

Swelling behavior was largely affected when these hydrogels were swollen in 5.0% NaCl at 40°C and pH 7.0 [Fig. 3(d)]. Decrease in the P_s values was observed as compared to the swelling observed in double distilled water. It suggests that the salt tolerance of these hydrogels is not high enough to swell in medium of high salt concentrations, yet it is quite appreciable. Increasing the ionic strength reduced the swelling degree because the low molecular weight ions (Na^+ and Cl^-) surrounded the charged groups and suppresses the swelling of the hydrogels.

The swelling behavior of the hydrogels was also studied in the presence of SDS. Because of the non-biodegradable nature of most of the surfactants, these are usually present as common effluents in most of the water bodies, hence their presence affects metal ion uptake. Depending upon the structure of the hydrogels, the presence of the surfactants increase[23] or decrease[24] water uptake level of the hydrogels near its critical micelle concentration (cmc), i.e., reflection of the capacity of the hydrogels to partition metal ions from the solution phase. Therefore, the swelling behavior of the hydrogels was studied in the presence of SDS using its 4.05, 8.32, and 12.15 mM/L at 40°C and pH 7.0 [Fig. 3(e)]. It is evident that for all the hydrogels maximum P_s was observed at 8.32 mM/L, i.e., near to the reported cmc of SDS at 40°C. It suggests that micellization occurs in the presence of SDS, hence, an increase in the water uptake at the near the cmc of the surfactant is obvious.

The partial hydrolysis of hydrogels by 0.5M NaOH enhances swelling by three to five times. Swelling was studied only at the optimum temperature, pH and [SDS], i.e., 40°C, 7.0, and 8.32 mM/L and 5.0% NaCl [Fig. 3(f–h)]. P_s of all the partially hydrolyzed crosslinked polymers increases as a function of time. It becomes almost constant after the initial increase. The increase in the swelling is due to the increase in the number of carboxylate groups in
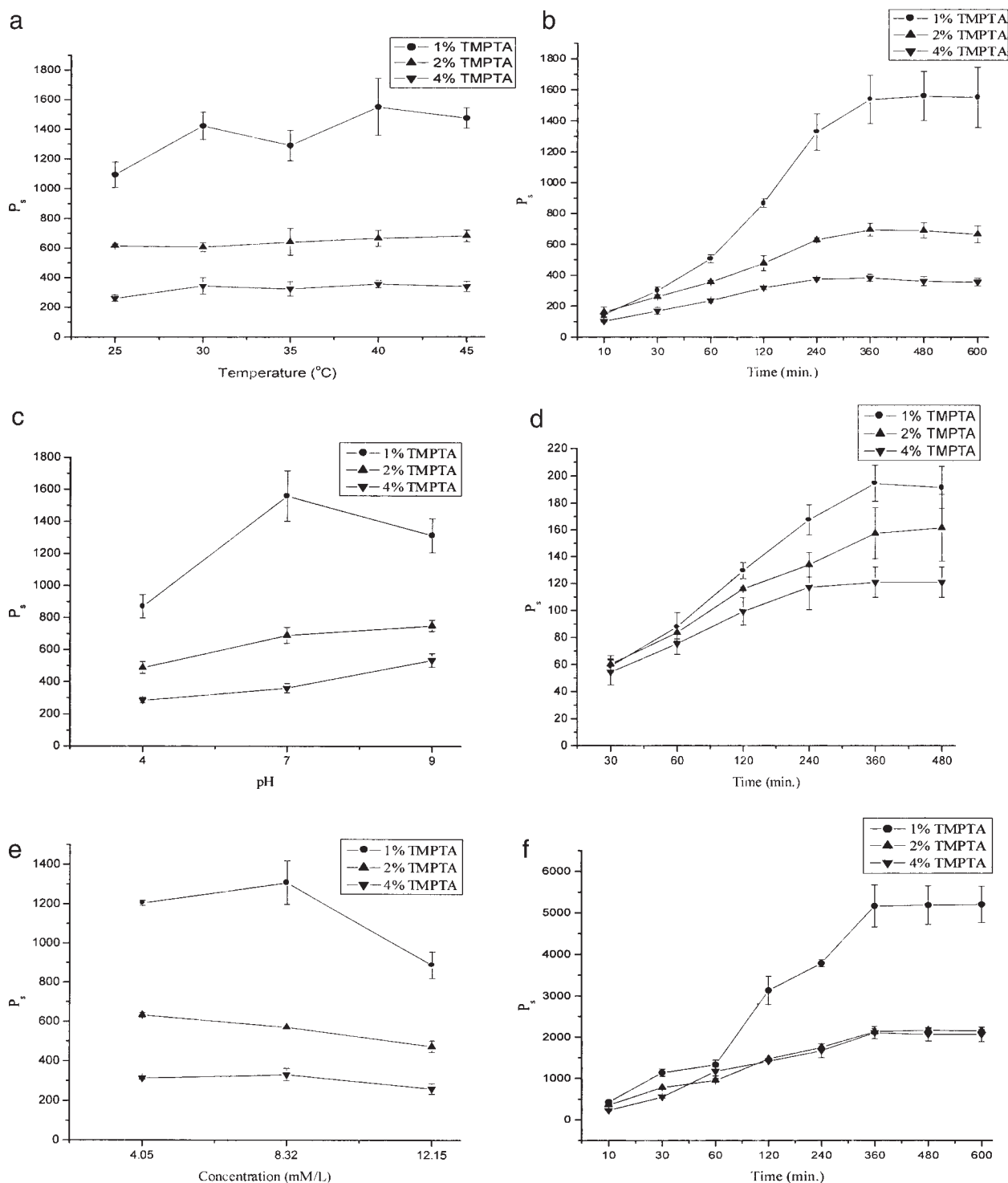
Figure 3 (a) P_s of poly(AAc-*co*-AAm)-*cl*-TMPTA hydrogels as a function of temperature (time = 600 min). (b) P_s of poly (AAc-*co*-AAm)-*cl*-TMPTA hydrogels as a function of time at 40°C. (c) P_s of poly(AAc-*co*-AAm)-*cl*-TMPTA hydrogels as a function of pH at 40°C (time = 480 min). (d) P_s of poly(AAc-*co*-AAm)-*cl*-TMPTA hydrogels in 5% NaCl as a function of time at 40°C. (e) P_s of poly(AAc-*co*-AAm)-*cl*-TMPTA hydrogels as a function of [SDS] at 40°C (time = 480 min). (f) P_s of partially hydrolyzed poly(AAc-*co*-AAm)-*cl*-TMPTA hydrogels as a function of time at 40°C. (g) P_s of partially hydrolyzed poly(AAc-*co*-AAm)-*cl*-TMPTA hydrogels as a function of time at 40°C in 5%NaCl. (h) P_s of partially hydrolyzed poly (AAc-*co*-AAm)-*cl*-TMPTA hydrogels as a function of time at 40°C in 8.32 m*M* SDS. (i) Salt sensitivity (*f*) of poly(AAc-*co*-AAm)-*cl*-TMPTA hydrogels as a function of time at 40°C in 5%NaCl. (j) Salt sensitivity (*f*) of partially hydrolyzed poly (AAc-*co*-AAm)-*cl*-TMPTA hydrogels as a function of time at 40°C in 5%NaCl.
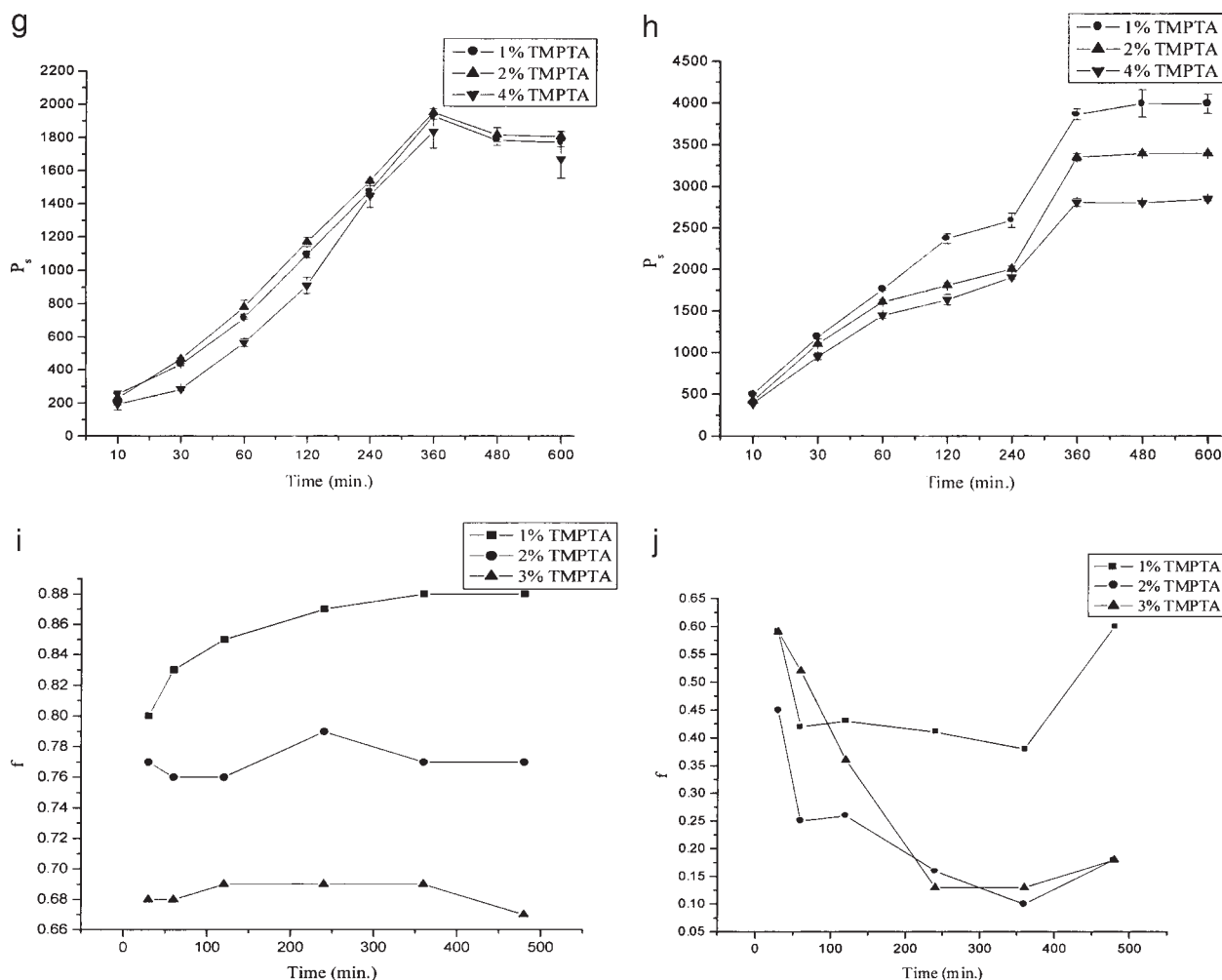
Figure 3 (*Continued from the previous page*)

the network. Swelling behavior of all the partially hydrolyzed hydrogels shows the common trends in swelling. The swelling of the hydrogels is significant in brine solution depicting the improvement in the salt tolerance of these hydrogels after hydrolysis, as compared to their precursors. Salt sensitivity of the partially hydrolyzed hydrogels and their precursors is compared [Fig. 3(i,j)]. The results of salt sensitivity are quite obvious. The precursor hydrogels are more salt sensitive than the partially hydrolyzed ones. The observation is important to assign end-uses to the hydrogels, as a hydrogel with the higher salt tolerance can be used in sea water.

Uranyl ions uptake studies

Hydrogels based on poly(AAc-*co*-AAm) have been used in metal ion uptake and enrichment studies.[6] However, the originality of the present study emanates form the fact that the use of hydrogels with TMPTA as crosslinker and post functionalization of the same by partial hydrolysis has not been reported

earlier. In the reported literature, the polymers are prepared by gamma ray initiation and uranyl ion adsorption has been studied without the use of NaCl. The use of NaCl was considered necessary to simulate the nature of sea water. These polymers are highly hydrophilic in nature, good absorbent of water even in the brine solution. These are thus capable of partitioning of metal ions from the metal ion solutions even in the presence of strong electrolytes like NaCl. Further, in polyelectrolytes numerous charges on polymer chains are created by the self ionization of poly(acid) and that create a higher electrostatic potential around them. Hence, in the present case, metal ion sorption is a combination of sorption due to the partitioning caused by the high water absorption capacity of the hydrogels, cation exchange at the carboxylate groups, especially after hydrolysis and also by simple adsorption on amide groups. Hence, understanding of the structural aspects of the hydrogels and their water uptake behavior provides important information to define their metal ion uptake properties.
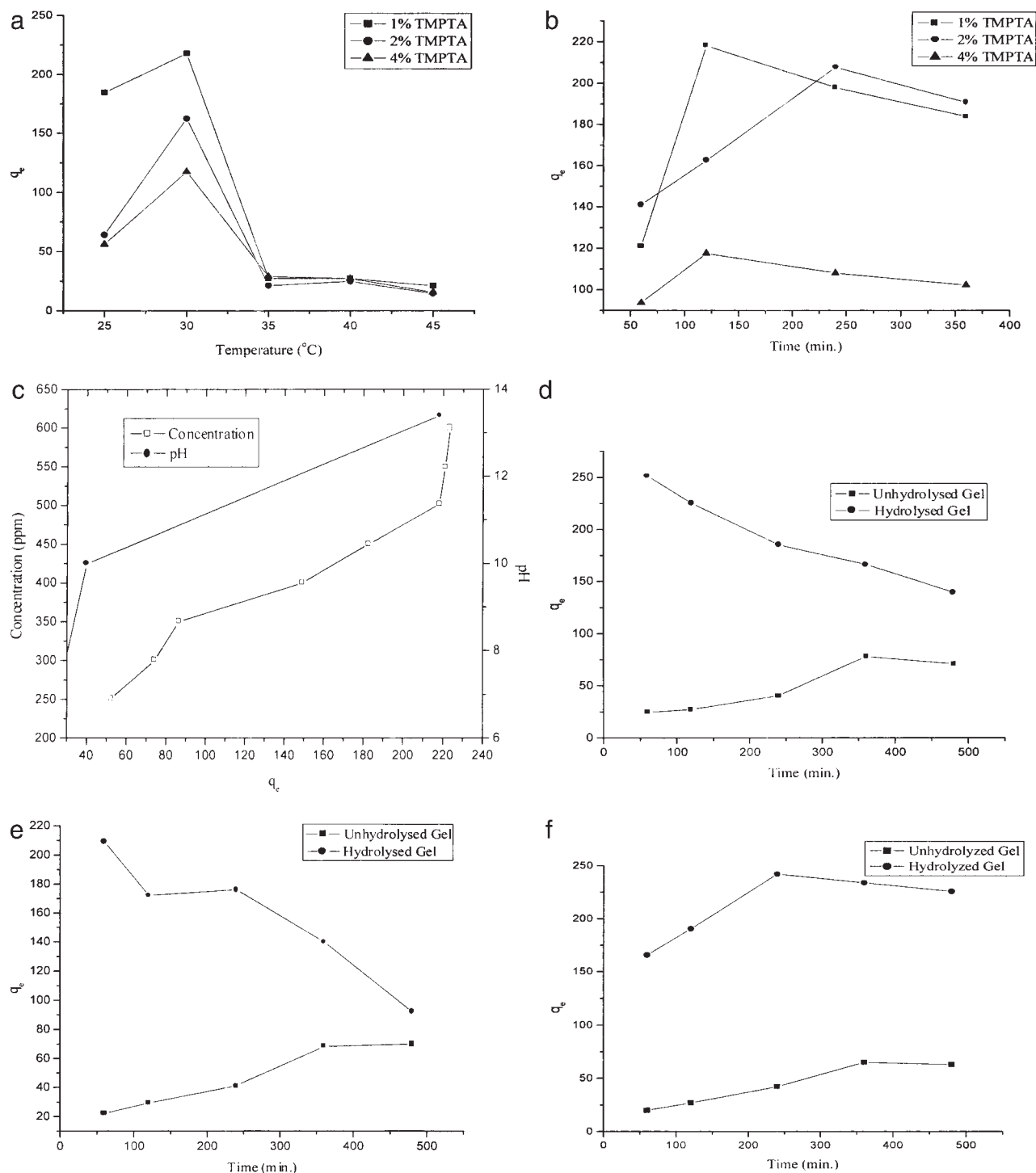
Figure 4 (a) q_e of poly(AAc-*co*-AAm)-*cl*-TMPTA hydrogels as a function of temperature in 502 ppm UN Solution (time = 120 min, pH = 13.4). (b) q_e of poly(AAc-*co*-AAm)-*cl*-TMPTA hydrogels as a function of time in 502 ppm UN Solution at 30°C (pH = 13.4). (c) q_e of poly(AAc-*co*-AAm)-*cl*-TMPTA hydrogels as a function of concentration in UN Solution(pH = 13.4) and as a function of pH in 502 ppm UN at 30°C (time =120 min). (d) q_e of poly(AAc-*co*-AAm)-*cl*-TMPTA (1%) hydrogels as a function of time in 502 ppm solution of UN at 30°C (pH = 7.0). (e) q_e of poly(AAc-*co*-AAm)-*cl*-TMPTA (2%) hydrogels as a function of time in 502 ppm solution of UN at 30°C (pH = 7.0). (f) q_e of poly(AAc-*co*-AAm)-*cl*-TMPTA (4%) hydrogels as a function of time in 502 ppm solution of UN at 30°C (pH = 7.0).

Uranyl ions uptake on the series of poly(AAc-*co*-AAm)-*cl*-TMPTA and their partially hydrolyzed forms was studied as a function of time, temperature

and pH using 502 ppm (1 m*M*) solution of uranyl nitrate in Na$_2$CO$_3$ buffer and 5%NaCl [Fig. 4(a,b)]. It was also studied as a function of different concentra-

tion of uranyl nitrate (251–600 ppm) [Fig. 4(c)]. Percent uptake or q_e value was found to be maximum for 502 ppm solution of uranyl nitrate at 30°C and pH 13.4 while network prepared with 1% crosslinker exhibits maximum uptake. Hence, ion uptake is affected by the structure of the hydrogel or more precisely it is related to its water uptake capacity, and also to the external factors like time, temperature, and pH. An increase in the temperature from 30 to 45°C causes a decrease in the amount of the adsorbed UO_2^{2+} ions, indicating that the adsorption is an exothermic process. The decrease in the uranyl uptake with the increase in temperature implies that the contribution of the adsorption processes to the overall ion uptake is appreciable as the ions gets desorbed at the higher temperatures. Further, on increasing the ionic strength the hydrogels can more effectively partition the ions from the salt solutions.

In the case of partially hydrolyzed hydrogels, q_e value increases as compared to their precursors, i.e., unhydrolyzed hydrogels [Fig. 4(d–f)]. The generation of the carboxylate groups after partial hydrolysis increase partitioning of the metal ions through enhanced water increase, and also due to the ion exchange of the UO_2^{2+} at the carboxylate groups. The uranyl uptake on these hydrogels is 255 mg g^{-1} of dry hydrogel from the uranyl nitrate solution in 5% NaCl which are comparatively better than the reported results in the literature.[6,25]

Evidence of uranyl ions uptake on the hydrogels is provided by FTIR of the hydrogels loaded with uranyl ions. On comparison of the spectra of the precursor and the uranyl ions loaded hydrogel, it has been observed that there are sharp differences in the two spectra (Fig. 2). A shift in the C=O stretching of acid and amide group of the hydrogel has been observed. Initially the peak of C=O stretching was observed at 1741.3 and 1653.6 cm^{-1} while in loaded hydrogel it was at 1664.4 and 1564.4 cm^{-1}. Similarly, N-H stretching of amide is shifted from 3446.4 to 3430 cm^{-1}. This indicate strong interaction of uranyl ion with C=O and N—H groups of the hydrogels. Additional peaks around 900 cm^{-1} can also be seen which are characteristics of O=U=O stretching.[26]

CONCLUSIONS

From the forgone discussion, it can be concluded that the hydrogels synthesized are environmentally sensitive as these respond to changes in the stimuli like temperature, pH, and the nature of the medium. These hydrogels also show salt tolerance, especially

after these were subjected to partial hydrolysis. From the uranyl uptake study, it is evident that the hydrogels are efficient candidates for the uranyl enrichment and its removal from water including from simulated sea water. The uranyl uptake capacity of these hydrogels is appreciable as 1 g of dry hydrogel sorbed up to 255 mg of uranyl ions from the uranyl nitrate solution. The uranyl uptake is temperature dependent as it decreased with an increase the temperature.

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