Superabsorbent Polymeric Materials VIII: Swelling Behavior of Crosslinked Poly[Sodium Acrylate-*co*-Trimethyl Methacryloyloxyethyl Ammonium Iodide] in Aqueous Salt Solutions

WEN-FU LEE, GU-HSI LIN

Department of Chemical Engineering, Tatung University, Taipei, Taiwan, Republic of China

ABSTRACT: A series of xerogels based on sodium acrylate (SA), cationic comonomer, trimethyl methacryloyloxyethyl ammonium iodide (TMMAI), and *N,N*-methylene-bis-acrylamide (NMBA) were prepared by inverse suspension polymerization. The water absorbency and the swelling behavior for these high absorbent polymers in deionized water and various saline solutions were investigated. Results indicated that the water absorbency for the present copolymer gel increased when a small amount of TMMAI monomer was introduced into the SA gel, then decreased with increase in TMMAI content. The water absorbency was 583 g H_2O/g for a gel sample in deionized water containing 2.5×10^{-3} molar fraction TMMAI. But a contrary result was observed for initial absorption rate, that is, the initial absorption rates increased with an increase of TMMAI in deionized water and 0.9 wt % NaCl solution. The absorbency in the chloride salt solution decreased with an increase in the ionic strength of the salt solution. Finally, the adsorption of copper ion by these gels was also investigated. © 2000 John Wiley & Sons, Inc. J Appl Polym Sci 79: 1665–1674, 2001

Key words: superabsorbent; water absorbency; swelling behavior; xerogel; sodium acrylate; inverse suspension polymerization

INTRODUCTION

Superabsorbent polymers were first reported by the U.S. Department of Agriculture, and much attention was paid to their applications. Because of their excellent characteristics, these polymers are widely used in many fields, such as disposable diapers, feminine napkins, soil for agriculture and horticulture, gel actuators, water-blocking tapes, medicine for drug delivery systems and absorbent pads, etc.[1–8] In such applications, water absorbency or water retention is essential. Hence, some researchers attempted to improve these absorbent polymers to enhance their absorbency, gel strength, and initial absorption rate.[9–23]

Flory explained the swelling mechanism for hydrophilic polymers containing a nonionic or ionic network structure.[24] Ogawa et al. studied the kinetics of the swelling of poly(sodium acrylate)[poly(SA)] by picture analysis with a microscope-VTR system, calorimetry, and gravimetry techniques.[25]

A series of superabsorbent polymers, for studying swelling behavior in deionized water and various saline solutions, were reported in our previ-

ous reports, such as poly(SA) containing nonionic comonomer 2-hydroxyethyl methacrylate (HEMA);[26] zwitterionic monomer 3,3-dimethyl methacryloyloxyethyl ammonium propane sulfonate (DMAPS),[27] or 3,3-dimethyl acrylamidopropyl ammonium propane sulfonate (DMAAPS);[28] anionic monomer sodium 2-acrylamido-2-methyl propane sulfonate (NaAMPS);[29] and poly(SA) with various initial total monomer concentrations.[30]

According to our previous studies, the SA gels containing nonionic comonomer HEMA had reduced water absorbency but improved initial absorption rate. However, contrary results were obtained from those gels containing the zwitterionic comonomer DMAPS, DMAAPS, and anionic comonomer NaAMPS. In addition, the water absorbency was decreased when the initial total monomer concentration of poly(SA) increased, but the opposite result for initial absorption rate was obtained. The poly(SA) gels containing cationic comonomer were not found in the previous literature, so a series of superabsorbent gels based on SA and TMMAI were prepared in this work. The water absorbency and swelling behavior for these xerogels in deionized water and various saline solutions were investigated.

EXPERIMENT

Materials

The materials used were purchased from Tokyo Kasei Industries Ltd., including acrylic acid (AA), N,N'-methylene-bis-acrylamide (NMBA). Sodium hydroxide and NMBA were used directly. 4,4′-Azobis(4-cyanovaleric acid) (ACVA) as an initiator and sorbitan monostearate (Span 60), as an inverse suspension stabilizer, were also purchased from Tokyo Kasei Industries Ltd. Methanol and cyclohexane were of analytical grade.
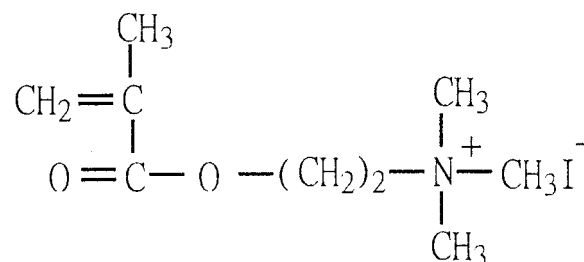
Preparation of SA Monomer Solution

The SA monomer was prepared as reported previously.[26]

Synthesis of TMMAI Monomer

The monomer trimethyl methacryloyloxyethyl ammonium iodide (TMMAI) was prepared as reported previously.[31] Yield: 92%, m p.: 184°C.

The structure is given below:

Inverse Suspension Polymerization

A 300-mL four-neck separable flask equipped with a reflux condenser, a stirring rod, and a thermometer was charged into 60-mL cyclohexane and 0.15 g sorbitan monostearate. The mixture was stirred until the sorbitan monostearate was dissolved (continuous phase).

The crosslinking agent, NMBA, and the appropriate amounts of TMMAI monomer, were introduced into SA monomer solution and the mixture was stirred until the NMBA was dissolved completely. The monomer solution and 0.15 g initiator, ACVA (dispersion phase), were introduced into the reactor. Air was flushed from the reactor by introducing nitrogen until the entire process was completed. The stirrer speed was maintained at 250 rpm. The polymerization was set at 70°C for 4 h. After the reaction was completed, the suspension solution was cooled and then precipitated by 800 mL cold methanol under stirring. The product was filtered and washed several times by the mixture of water and methanol (1 :9 vol). The product was then dried in a vacuum oven at 70°C for 1 day. A white powdered polymer was obtained, and the yield was over 95%. The compositions of samples are listed in Table I.

Measurement of Properties

The samples were dried in a vacuum oven at 70°C before any tests. All of the samples were used with the particle size in the range of 60 and 100 mesh.

Saturated Absorbency

The sample (50 mg) was immersed in an excess of deionized water or 0.9 wt % NaCl(aq) for at least 8 h to reach the swelling equilibrium at room temperature.

Suction Filtration Method

When the sample reached swelling equilibrium, the residual water was removed by suction filtra-

Table I   Feed Compositions of SA/TMMAI Xerogels

Sample No.	SA g	TMMAAI g (mol %)	NMBA g (mol %)	Yield (%)	Water Soluble (%)
AP1	20	0	0.07 (2.14×10^{-3})	98.8	8
AP2	19.95	0.16 (2.5×10^{-3})	0.07 (2.14×10^{-3})	96.7	6
AP3	19.85	0.48 (7.5×10^{-3})	0.07 (2.14×10^{-3})	98.6	8
AP4	19.7	0.96 (1.5×10^{-2})	0.07 (2.14×10^{-3})	94.8	7
AP5	19.55	1.43 (2.25×10^{-2})	0.07 (2.14×10^{-3})	98.6	12
AP6	19.4	1.91 (3.0×10^{-2})	0.07 (2.14×10^{-3})	99.1	11
AP7	19.4	1.91 (3.0×10^{-2})	0.05 (1.53×10^{-3})	97.5	9
AP8	19.4	1.91 (3.0×10^{-2})	0.10 (3.06×10^{-3})	98.4	6
AP9	19.4	1.91 (3.0×10^{-2})	0.15 (4.60×10^{-3})	96.8	6
AP10	19.4	1.91 (3.0×10^{-2})	0.20 (6.12×10^{-3})	97.8	11

The total weight of water is 60 g, ACVA, 0.1 g; span 60, 0.15 g; and cyclohexane.

tion with an aspirator (250 mmHg) for 5 min. The gel was weighed, and the equilibrium absorbency Q_{eq} was calculated by the following equation:

$$Q_{eq} = \frac{W_{wet} - W_{dry}}{W_{dry}} \quad (1)$$

where W_{dry} is the weight of the dried sample, and W_{wet} is the weight of the swollen sample.

"Tea Bag" Method

The tea bag was made of 250-mesh nylon screen. The tea bag containing the sample (50 mg) was immersed entirely in deionized water or the saline solution and kept there until the swelling equilibrium was reached, and then hung up for 15 min to drain the excess solution and weighed. The equilibrium absorbency was calculated by eq. (1).

Water Absorbency in Various Saline Solutions

The dried sample (50 mg) was immersed in an excess of various saline solutions with different concentrations [LiCl(aq), NaCl(aq), KCl(aq), MgCl$_2$(aq), SrCl$_2$(aq), BaCl$_2$(aq), CuCl$_2$(aq), FeCl$_3$(aq)] and remained there for at least 8 h. The sample was filtered with suction and weighed. The water absorbency in the above saline solution could be obtained.

Kinetics of Swelling

A technique based on the demand wettability (*DW*) method was adopted.[16] A graduated burette with an air inlet at the bottom was linked by a flexible tube to a plate covered with a metallic grid (250 mesh). After filling with the saline so-

lution, the burette was closed at the top, and air was introduced while the xerogel absorbed water. The sample (50 mg) of absorbent was placed on the grid, and the swelling kinetics were easily measured. The absorbency at every time interval was calculated by following eq. (2):

$$Q = \frac{V_s \times d_{H_2O}}{W_d} \quad (2)$$

where Q is the characteristic absorbency, V_s is sorbed volume, d_{H2O} is the density of water, and W_d is the weight of dry sample.

Measurement of Ions Adsorption from Salt Solutions

The dried sample (50 mg) was immersed in 50 mL of various concentrations (C_0) of CuCl$_2$(aq) solution to absorb and remain there until equilibrium. The sample was filtered and the residue volume (V_1) was measured. The residual concentration of the copper ion (C_1) in aqueous solution was analyzed at 815 nm by UV-spectrophotometer (JASCO V530). The adsorbed amount was calculated by following formula:

$$AD = \frac{C_0 \times 0.05 - C_1 \times V_1}{W_d} \quad (3)$$

where AD is the adsorbed amount of the copper ion and W_d is the weight of the dried sample.

RESULTS AND DISCUSSION

The swelling behavior of the superabsorbents depends on the nature of the polymer and the char-
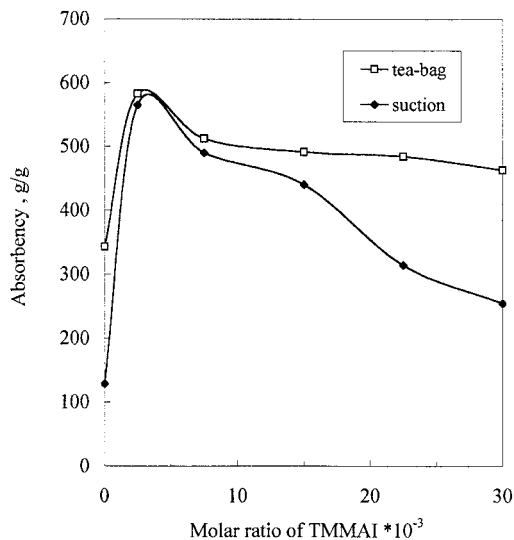
Figure 1 Effect of TMMAI content on water absorbency in deionized water.

acteristics of the external solution. The polymer's nature involves the nature of the charge, ionic content, and crosslinking density. The characteristics of the external solution include the charge number and ionic strength.

The swelling behaviors of SA series superabsorbents were studied in our previous articles.[26–32] A series of SA/TMMAI superabsorbent polymers is discussed in this article. The effect of TMMAI content on water absorbency and initial absorption rate was investigated.

Effect of TMMAI Content on Saturated Absorbency

The effect of TMMAI content for SA/TMMAI copolymeric gels on water absorbency in deionized water measured by the tea bag method and the suction method is shown in Figure 1. The results indicate that the water absorbency of the present copolymeric xerogels is larger than that of poly(SA) (AP1), and the water absorbency decreases with increasing of the molar ratio of TMMAI(A2–A6). According to the Flory's swelling theory,[24]

$$Q^{5/3} = [(i/2V_uS^{1/2})^2 + (1/2 - \chi_1)/v_1]/(\nu_e/V_o) \quad (4)$$

where i/V_u is the concentration of fixed charge referred to unswollen network, S is ionic strength of the external solution, $(1/2 - \chi_1)/v_1$ is related to the affinity of the gel for water, ν_e/V_o is number of effective chains per unit volume of swollen poly-

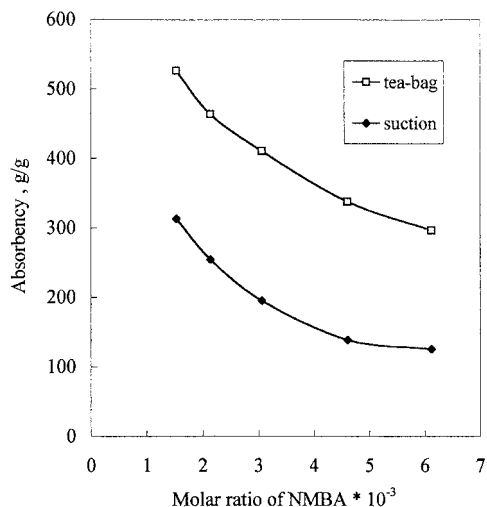mer, which is related to the crosslinking density of the gel.

Hence, the water absorbency of the gel is dependent on ionic osmotic pressure, crosslinking density, and the affinity of the gel for water. The crosslinking density of SA/TMMAI copolymeric gels and the ionic strength in the external solution were fixed. The water absorbency for the gel is dependent on the concentration of fixed charge in the gel and the affinity of the gel for water. When the cationic monomer TMMAI is introduced into the copolymeric gel, the iodide ion (I⁻) was dissociated and the quaternary ammonium group (R_4N^+) with positive charge was formed. The affinity of the quaternary ammonium group (R_4N^+) is stronger than that of the carboxylate group (COO^-). Therefore, the water absorbency of the present gel is higher than that of poly(SA) (AP1) when the cationic monomer TMMAI is introduced into the copolymeric gel.

However, the quaternary ammonium group (positive charge) would bind with the carboxylate group (negative charge), and therefore, the fixed charge concentration of the polymer network decreases. This behavior reduces the negative charge repulsion of the polymer network and the water absorbency of the gel decreases. This behavior was also observed from our previous study.[33]

Furthermore, the water absorbency in deionized water measured by the tea bag method and the suction method are 463–583 and 254–565 g water/g dry sample, respectively. From these data, we can find that the water absorbency, measured by the tea bag method, is larger than that measured by the suction method. This is because the tea bag method cannot remove the free water from the gel particles. Similar results were observed in our previous work.[27, 29,30]

Effect of the Extent of Crosslinking Agent on Water Absorbency

The effect of the extent of crosslinking agent on water absorbency for SA/TMMAI copolymeric gels is shown in Figure 2. The results in Figure 2 indicate that with more crosslinking agent the water absorbency is lower. The copolymeric gel containing 3.0×10^{-2} molar ratio of TMMAI exhibits water absorbency of 313 g water/g sample when the molar ratio of NMBA is 1.53×10^{-3} (AP7), but the water absorbency decreases to 125 g water/g dry sample when the molar ratio of NMBA is 6.12×10^{-3} (AP10). This phenomenon

Figure 2 Effect of the extent of a crosslinking agent on absorbency in deionized water.

Figure 3 Water absorbency of poly(SA-TMMAI) gel in NaCl aqueous solution with various ionic strengths.

conforms to the Flory theory; the water absorbency is also related to the elastic force between the polymer chains. With more crosslinking agent, the higher the crosslinking density, the stronger the elastic-force of the polymer chain, and the lower the water absorbency. This occurrence also corresponds to our previous study for poly(SA-*co*-NaAMPS).[30]

Effect of Salt Solution on Water Absorbency

To investigate the effect of saline solution on water absorbency for the present copolymeric gels in various salt solutions, samples AP2 and AP5 were chosen. The water absorbencies for samples AP2 and AP5 in various saline solutions with different cations containing common anion were studied.

Water Absorbency in Various Salt Solutions

The water absorbency for the present poly(SA-*co*-TMMAI) copolymeric gels in NaCl(aq), CuCl$_2$(aq), and FeCl$_3$(aq) are shown in Figures 3, 4, and 5, respectively. The water absorbencies for samples AP2 and AP5 in different valence salt solutions are shown in Figures 6 and 7, respectively.

The results shown in these figures indicate that the water absorbency for the copolymeric gels in various salt solutions decreases with increasing ionic strength of salt solution. This can be attributed to the cations in the water, such as the sodium ion (Na$^+$), the cupric ion (Cu^{2+}), and the ferric ion (Fe^{3+}) in this article. The cation

would neutralize the carboxylate group (COO$^-$). Based on the Donnan membrane equilibrium theory, the difference in the ionic osmotic pressure between the gel and the external solution decreases as the ionic strength of the salt solution increases. Therefore, the water absorbency decreases when the ionic strength in the external solution increases according eq. (4) (see Figs. 3–7).

Moreover, the swelling curves for divalent (Fig. 4) and trivalent (Fig. 5) salt solutions are steeper than those for monovalent (Fig. 3) salt solutions. The water absorbency converged to zero when those gels were immersed into the concentrated divalent and trivalent salt solutions (10^{2-} M), but that did not occur when those gels were immersed
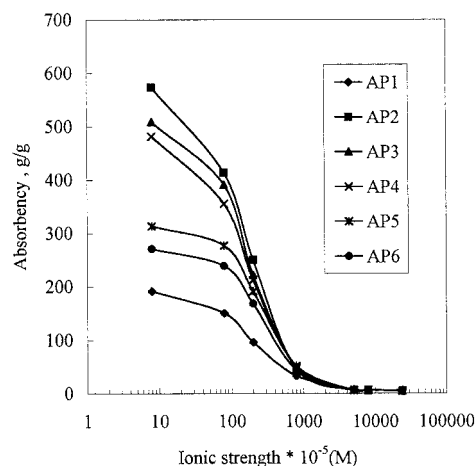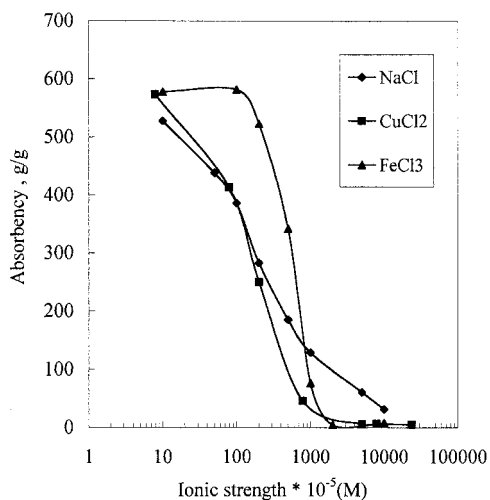

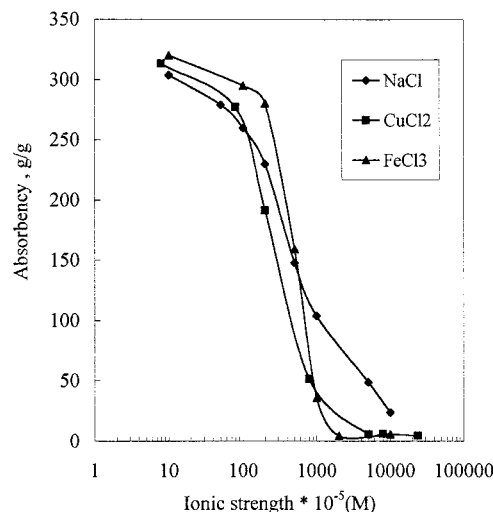**Figure 4** Water absorbency of poly(SA-TMMAI) gel in CuCl$_2$ aqueous solution with various ionic strengths.
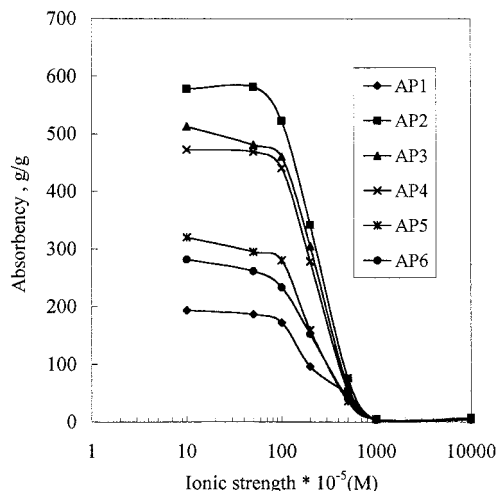
Figure 5 Water absorbency of poly(SA-TMMAI) gel in $FeCl_3$ aqueous solution with various ionic strengths.

into the monovalent salt solutions. This is because the divalent cupric ion (Cu^{2+}) and trivalent ferric ion (Fe^{3+}) would form complexes with the carboxylate group. Hence, the swelling curves apparently decrease at the gel transition concentration, and the water absorbency converges to zero in concentrated salt solutions (complexing effect).

On the other hand, the tendency of water absorbency for the gel is in the order of $Fe^{3+} > Cu^{2+} > Na^+$ in the dilute solution (10^{3-} M). In other words, the influence of monovalent, divalent, and trivalent cations with common anion (Cl^-) on water absorbency shows a tendency of trivalent > di-

Figure 6 Water absorbency of sample AP2 in NaCl, $CuCl_2$, and $FeCl_3$ aqueous solution with various ionic strengths.

Figure 7 Water absorbency of sample AP5 in NaCl, $CuCl_2$, and $FeCl_3$ aqueous solution with various ionic strengths.

valent > monovalent cations in the dilute solution. The result is related to the charge number of the ion. This occurrence makes the hydration ability become stronger, and the water absorbency increases. Similar results were observed from our previous reports[27-31] and conform to the formation constant of complexation.

Effect of Salt Solutions with Different Cations with a Common Anion (Cl^-) Salt Solution

The water absorbencies for sample AP2 in monovalent and divalent cations with a common anion (Cl^-) salt solutions are shown in Figures 8 and 9, respectively. The results show that the water absorbency for the copolymeric gel in monovalent and divalent cations salt solution is in the order of $LiCl > NaCl = KCl$ (Fig. 8) and $MgCl_2 > SrCl_2 = BaCl_2$ (Fig. 9), respectively. These results imply that the smaller the cationic radius, the higher the water absorbency. This is because the smaller the cationic radius, the stronger the hydration ability of the cation, that is, the binding ability to the carboxylate group is weakened and leads to the water absorbency increase.
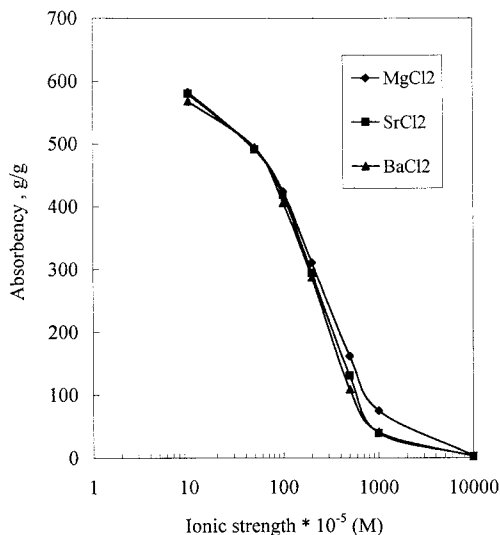
Effect of TMMAI Content on Absorption Rate for SA/TMMAI Copolymeric Gels

The swelling kinetics of spherical polyacrylamide absorbents were described in the model by Tanaka and Fillmore,[34] and was generalized by
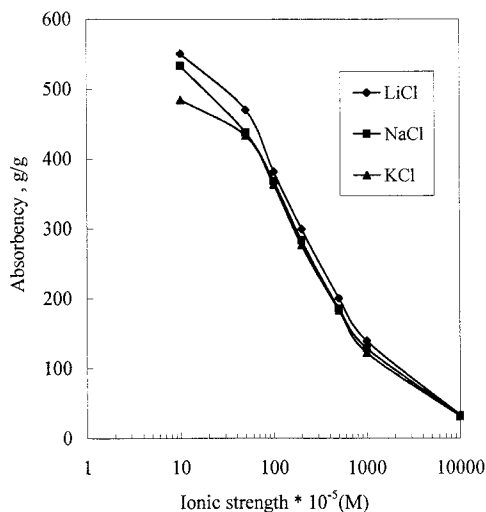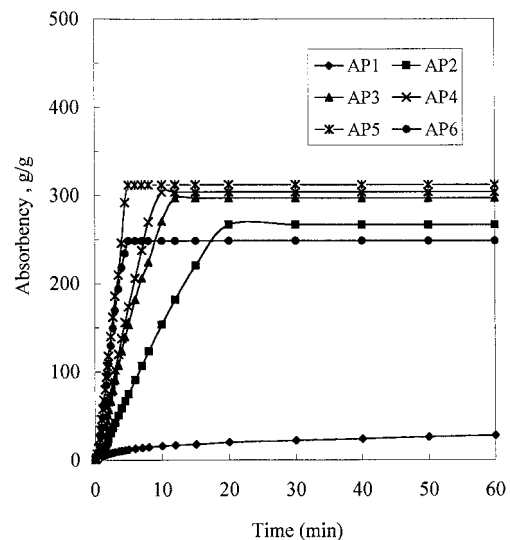
Figure 8 Water absorbency of sample AP2 in LiCl, NaCl, and KCl aqueous solution with various ionic strengths.

Figure 10 Absorption rate in deionized water for Poly(SA-TMMAI) xerogel measured by the DW method.

Candau et al.,[35] who considered a single isolated gel sphere partly swollen and put in an excess of solvent. There is an osmotic pressure difference, and the gel swells to reach a new equilibrium volume. The swelling behavior of the absorbent was explained by water penetrating into the polymeric network. Buchanan[36] has suggested that the swelling kinetic for the absorbent is significantly influenced by swelling capacity, size distribution of powder particles, specific surface area, and apparent density of the polymer.
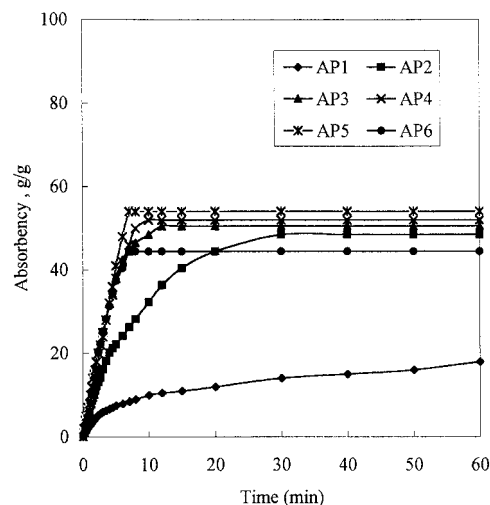
Figures 10 and 11 show the absorption rate for a series of poly(SA-*co*-TMMAI) gels in deionized water and 0.9 wt % NaCl(aq). The results shown in these figures indicate that the initial absorption rate rapidly increases with increasing content of TMMAI. This is due to the cationic TMMAI monomer introduced into the copolymeric gel, the iodide ion was dissociated and the quaternary ammonium group (R_4N^+) was formed. The affinity of the quaternary ammonium group in the cationic TMMAI monomer introduced into the copolymeric gel would enhance the absorption

Figure 9 Water absorbency of sample AP2 in $MgCl_2$, $SrCl_2$, and $BaCl_2$ aqueous solution with various ionic strengths.

Figure 11 Absorption rate in 0.9 wt % NaCl for Poly(SA-TMMAI) xerogel measured by the DW method.

Table II   The Absorption Characteristics for SA Gel Systems in Deionized Water and 0.9 wt % NaCl Aqueous Solution

Conditions	AP1	AP2	AP3	AP4	AP5	AP6
			Deionized water			
Initial absorption rate (g/min)						
(30 s)	4.0	8.1	20.2	24.0	28.0	20.2
(1–3 min)	2.6	16.2	34.3	36.0	70.0	66.7
t_{req} (min)	300	20	12	10	5	5
Q^*_{eq} (g H$_2$O/g)	50	267	297	304	312	248
			0.9 wt % NaCl			
Initial absorption rate (g/min)						
(30 s)	3.0	6.0	6.1	8.0	10.0	6.1
(1–3 min)	1.7	4.5	8.0	8.0	8.0	9.6
t_{req} (min)	100	40	8	12	7	7
Q^*_{eq} (g H$_2$O/g)	21	48	51	52	54	44

t_{req}: time require to approach equilibrium.
Q^*_{eq}: equilibrium water absorbency by DW method.

rate for the copolymeric gel. Moreover, the initial absorption rates for these gels, for the initial 30-s period in 0.9 wt % NaCl(aq), is slower than that in deionized water (see Table II). This is primarily attributed to the reduction of the difference in ionic osmotic pressures between the gel and the external solution.

According to Yao and Zhou,[37] the swelling rate can be described by the following equation:

$$-\log(Q_{eq} - Q) = K/2.303T \qquad (5)$$

where Q_{eq} is the equilibrium absorbency, Q is the characteristic absorbency, K is the swelling kinetic constant, and T is the characteristic swelling time. From eq. (5), T is defined at $Q = 0.632$ Q_{eq}. Table III presents the influences of the content of TMMAI on T, K, and Q of the Poly(SA-co-

TMMAI) absorbent in deionized water and in the saline solution. Table III reveals that T decreases and Q and K increases with increasing the content of TMMAI. The values of K for the copolymeric gels in deionized water are larger than those in the saline solution.

Adsorption of Copper Ion

Figure 12 shows the adsorbed amount of copper ion (Cu^{2+}) by AP1 and AP3 gels in various concentrations of $CuCl_2$(aq). The result shows that the more the carboxylate group content in the gel, the stronger the adsorptive ability (AP1 > AP3) [Fig. 12(a)]. The adsorption of copper ion is primarily attributed to the number of carboxylate groups in the gel, that is, with more carboxylate groups in the gel, the higher the amount the cop-

Table III   Absorption Characteristics for Poly(SA-TMMAI) Gel Systems on the Characteristic Swelling Time (T), the Swelling Kinetic Constant (K), and the Characteristic Absorbency (Q) in Deionized Water and 0.9 wt % NaCl Aqueous Solution

Sample No.	T (min)		K (min^{-1})		Q (g H$_2$O/g Dried Sample)	
	H$_2$O	0.9 wt % NaCl	H$_2$O	0.9 wt % NaCl	H$_2$O	0.9 wt % NaCl
AP1	84	26.4	0.035	0.078	31.6	13.3
AP2	11.03	9.3	0.416	0.309	168.5	30.6
AP3	6.24	4.0	0.752	0.733	187.7	31.9
AP4	5.57	4.2	0.847	0.699	192.1	32.9
AP5	3.23	4.3	1.469	0.700	197.2	34.1
AP6	2.79	3.5	1.619	0.796	157	28.1
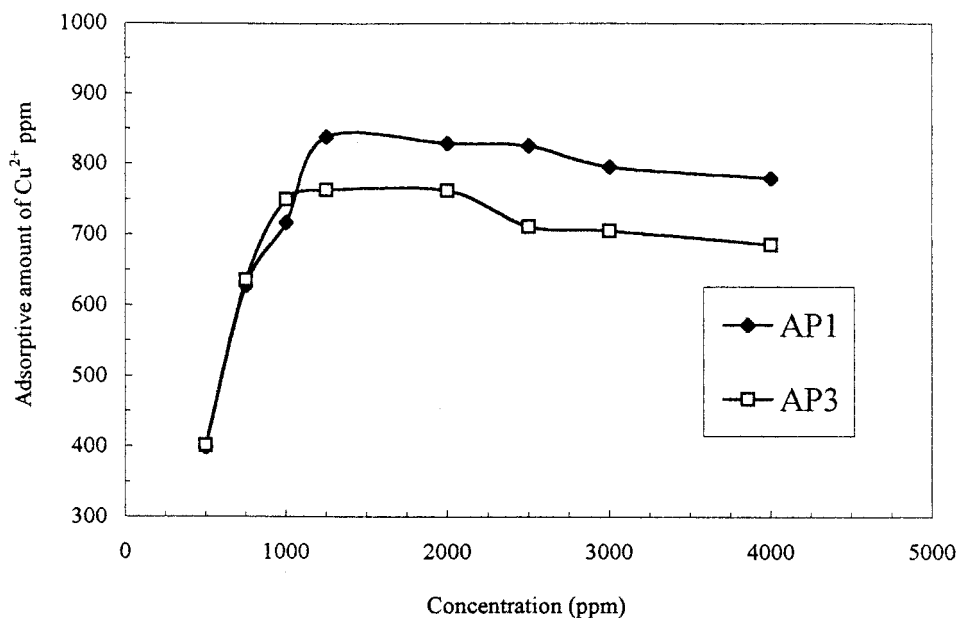
(a)
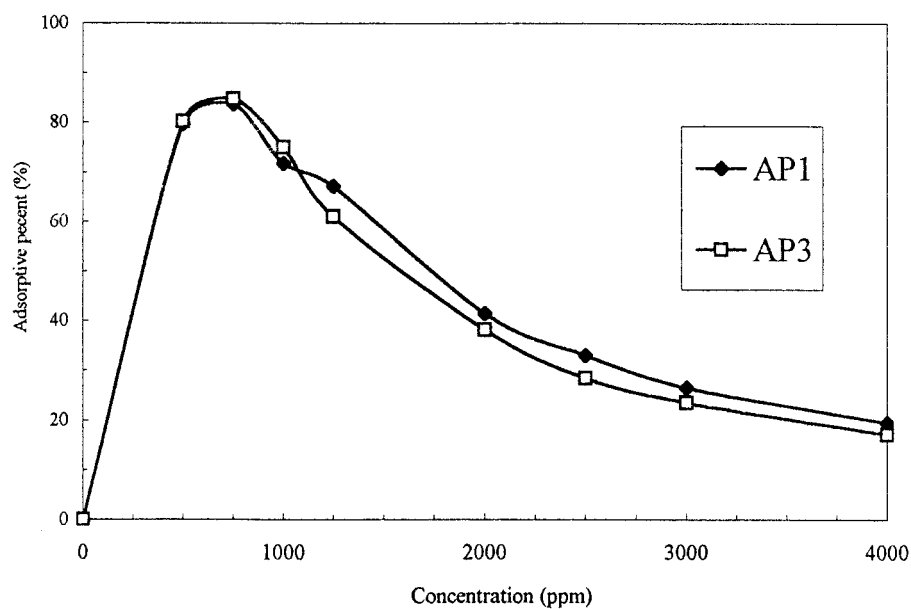
(b)

Figure 12   Adsorptive amount Cu^{2+} in various $CuCl_2$ concentrations for sample AP1 and AP3.

per ion adsorbed. This behavior conforms to the previous reports.[26,30,32] Furthermore, the optimum adsorbed percent (82%) for the present absorbent appears when the original copper concentration is 750 ppm [Fig. 12(b)].

CONCLUSION

The swelling behavior of the superabsorbents prepared from SA and TMMAI indicated that the water absorbency and initial absorption rate were

improved when a small amount of the TMMA2 monomer was introduced into the copolymeric gels. However, the greater the extent of crosslinking agent, the lower is the water absorbency.

The water absorbency for the copolymeric gels in various salt solutions decreases with increasing ionic strength of salt solution. The water absorbency converges to zero when these gels are immersed into the concentrated divalent and trivalent salt solutions ($>10^{-2}$ M), but that is not so when these gels are immersed into the monovalent salt solutions.

Finally, with more carboxylate groups in the gel, the stronger the ability for the gel to adsorb copper ion (Cu^{2+}). Furthermore, the optimum adsorbed percent of copper ion (Cu^{2+}) when the original copper concentration is 750 ppm.

REFERENCES

1. Buchholz, F. L. Superabsorbent Polymers Science and Technology; Buchholz, F. L.; Peppas, N. A., Eds.; ACS Symposium Series 573; American Chemical Society, Washington, DC, 1994.
2. Sakiyama, T.; Chu, C. H.; Fujii, T.; Yano, T. J Appl Polym Sci 1993, 50, 2021.
3. Yoshida, M.; Asano, M.; Kumakura, M. Eur Polym J 1989, 25, 1197.
4. Shiga, T.; Hirose, Y.; Okada, A.; Kurauchi, T. J Appl Polym Sci 1992, 44, 249.
5. Shiga, T.; Hirose, Y.; Okada, A.; Kurauchi, T. J Appl Polym Sci 1993, 47, 113.
6. Hogari, K.; Ashiya, F. Advances in Superabsorbent polymers; American Chemical Society: Washington, DC, 1994.
7. Ericksen, P. H.; Nguyen, H. V.; Oczkowski, B.; and Olejnik, T. A. Eur. Pat. 40087 (1981).
8. Kobayashi, T. Kobunshi 1987, 36, 612.
9. Taylor, N. W.; Fanta, G. F.; Doane, W. M.; Russell, C. R. J Appl Polym Sci 1978, 22, 1343.
10. Burr, R. C.; Fanta, G. F.; Doane, W. M. J Appl Polym Sci 1979, 27, 2713.
11. Fanta, G. F.; Burr, R. C.; Doane, W. M.; Russell, C. R. J Appl Polym Sci 1979, 24, 1384..
12. Kejun, Y.; Berlian, W. J Appl Polym Sci 1990, 41, 3079.
13. Fanta, G. F.; Burr, R. C.; Doane, W M. J Appl Polym Sci 1979, 24, 2015.
14. Yoshinobu, M.; Morita, M.; Sakata, I. J Appl Polym Sci 1992, 45, 805.
15. Lokhande, H. T.; Varadarjan, P. V.; Iyer, V. J Appl Polym Sci 1992, 45, 2031.
16. Castel, D.; Ricard, A.; Audebert, R. J Appl Polym Sci 1990, 39, 11.
17. Zoda, I. Funct Mater 1986, 6, 76.
18. Isomi, K. Jpn Kokai, Tokyo Koho JP, 56, 707 (1989).
19. Nagasuna, K.; Suminaga, N.; Kimura, K.; Shimonura, T. Jpn Kokai, Tokyo Koho JP, 126, 234 (1989).
20. Imada, H.; Fujiwaka, M. Jpn Kokai, Tokyo Koho JP, 141, 938 (1989).
21. Fujio, A.; Komae, T.; Yutaka, Y. Jpn Kokai, Tokyo Koho JP, 210, 463 (1989).
22. Yada, S.; Shibano, T.; Ito, K. Jpn Kokai, Tokyo Koho JP, 215, 801 (1990).
23. Sano, M.; Mikamo, H.; Suehiro, T.; Wakabayashi, N. Jpn Kokai, Tokyo Koho JP, 258, 839 (1991).
24. Flory, P. J. Principle of Polymer Chemistry; Cornell University Press: Ithaca, NY, 1953.
25. Ogawa, I.; Yamano, H.; Miyogawa, K. J Appl Polym Sci 1993, 47, 217.
26. Lee, W. F.; Wu, R. J. J Appl Polym Sci 1996, 62, 1099.
27. Lee, W. F.; Wu, R. J. J Appl Polym Sci 1997, 64, 1701.
28. Lee, W. F.; Yeh, P. L. J Appl Polym Sci 1997, 66, 499.
29. Lee, W. F.; Yeh, P. L. J Appl Polym Sci 1997, 64, 2371.
30. Lee, W. F.; Hsu, C. H. J Appl Polym Sci 1998, 69, 229.
31. Lee, W. F.; Tsai, C. C. Polymer 1994, 35, 2210.
32. Lee, W. F.; Tu, Y. M. J Appl Polym Sci 1999, 72, 1221.
33. Lee, W. F.; Huang, Y. L. J Appl Polym Sci, to appear.
34. Tanaka, T.; Fillmore, D. J. J Chem Phys 1979, 70, 1214.
35. Candau, S. J.; Bastide, T.; Delsanti, M. Adv Polym Sci 1982, 41, 27.
36. Buchanan, K. J.; Hird, B.; Letcher, T. M. Polym Bull 1986, 15 325.
37. Yao, K. J.; Zhou, W. J. J Appl Polym Sci 1994, 53, 1533.