Poly(sulfobetaine)s and Corresponding Cationic Polymers. VIII. Synthesis and Aqueous Solution Properties of a Cationic Poly(methyl iodide quaternized styrene–*N,N*-dimethylaminopropyl maleamidic acid) Copolymer

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ABSTRACT: A cationic poly(methyl iodide quaternized styrene–*N,N*-dimethylanipropylmaleamidic acid) copolymer was synthesized through amidoacidification reaction of styrene-maleic anhydride copolymer with *N,N*-dimethylaminopropylamine (ring-opening reaction). Its properties in various aqueous salt solutions and pH solutions were studied by measurements of reduced viscosity and intrinsic viscosity. The results indicated that the reduced viscosity and intrinsic viscosity of this cationic polyelectrolyte were related to the type and concentration of the added salts and the results also showed a contrary tendency in some salts with monovalent acid groups to polyelectrolyte. At the same time, some salt ions were observed to strongly attract the quaternary ammonium group of the cationic polymeric side chain and resulted in agglomeration of the polymers. © 2001 John Wiley & Sons, Inc. J Appl Polym Sci 80: 1619–1626, 2001

Key words: polyelectrolyte; styrene–*N,N*-dimethylaminopropyl maleamidic acid copolymer; reduced viscosity; intrinsic viscosity

INTRODUCTION

Cationic quaternary polyelectrolytes have been widely used in adhesive, coating, textile, hair conditioner, flocculant, and other related industries.[1–7] The functional groups on the side chain of the cationic polymers are usually ammonium (primary, secondary, tertiary, and quaternary amines), sulfonium, or phosphonium groups.[8] These cationic monomers with quaternary ammonium group are usually the quaternary ammonium group sites on the polymeric side chain.[8–18] The effect of various salt ions on the interaction of polyelectrolytes in the aqueous solution was investigated and the site-binding interaction of salt ions and polymer using Huggins equation k' was also investigated.[19–33] The counterion size was found to affect the degree of side binding for salt ion-attracting polymers. Cationic polyelectrolytes derived from styrene–maleic anhydride copolymer were reported on and proposed as flocculants by Isaacson and Young,[34] and Pratt and Diefenbach.[7]

A series of cationic polymers prepared from cationic monomers with different electrodrawing groups and by varying the methylene units between the charge groups, were researched previously in our laboratory such as poly(trimethyl acrylamido propyl ammonium iodide) [poly(TMAAI)], poly(methyl iodide quaternized styrene–dimethyl aminopropyl maleimide copolymer) [poly(MIQSD-MAPM)], and poly(methyl iodide quaternized acrylamide–*N,N*-dimethylaminopropyl maleimide copolymer [poly(MIQADMAPM)].[35–37] The previous

article on poly(TMAAI) showed that soft salt anions were more easily bound on the quaternary ammonium (R_4N) of poly(TMAAI) than those of hard salt anions.[35] In our MIQSDMAPM or MIQADMAPM copolymer synthesis, the reactive group (amine) was first attached to the styrene-maleic anhydride (SMA) or acrylamide-maleic anhydride copolymer framework, followed by a dehydration (ring-closing) reaction to obtain the styrene–N,N-dimethylaminopropylmaleimide (SDMAPM) or the acrylamide–N,N-dimethyl-aminopropylmaleimide (ADMAPM) which are tertiary amines, and then by a single reaction which generated the ionic structure.

The aqueous salt solution properties of MIQS-DMAPM and MIQADMAPM copolymers were studied in previous reports.[36,37] Hence, the investigation of aqueous solution properties for the methyl iodide quaternized styrene–dimethyl-aminopropylmaleamidic acid copolymer (MIQSD-MAPMA), especially the reduced viscosity and intrinsic viscosity in the presence of various salts, were the main interest in this research.

EXPERIMENTAL

Materials

Styrene, N,N-dimethyl amino propyl amine, dimethyl sulfoxide (DMSO), ethyl methyl ketone, and methyl iodide (MI) were reagent grade and used as received. Maleic anhydride (mp 56°C) and azobisisobutyronitrile were purified by recrystallization in methanol. Methanol and acetone were distilled twice. Deionized water had a resistivity of $18M\Omega$

Preparation of SMA Copolymer

Styrene (18.237 g, 0.175 mol), maleic anhydride (17.162 g, 0.175 mol), and ethyl methyl ketone (150 mL) were introduced into a 250-mL volume flask equipped with a stirrer, a cooler, and a thermometer. The contents were stirred at 80°C for complete dissolution. To the solution, azobisisobutyronitrile (0.45 wt % based on total monomers) as an initiator was added and the solution was stirred at the same temperature for 5.5 h under nitrogen atmosphere. The solution was then precipitated with diethyl ether. The product was then repeatedly dissolved and precipitated from acetone with diethyl ether, and then dried at 80°C for 24 h under vacuum to obtain the SMA

copolymer. The yield was 98%. The polymer was analyzed by elemental analysis. Elemental analysis for $C_{12}H_{10}O_3$: Calcd: C, 71.28%; H, 4.98%; O, 23.74%. Found: C, 70.78%; H, 5.04%; O, 24.18%.

Amidoacidification of SMA

SMA, 10.6 g, was mixed with 200 mL of acetone in a 500-mL single-necked flask at 55°C and stirred until it was completely dissolved. When the temperature cooled down to 40°C, 6.75 g of N,N-dimethyl amino propyl amine mixed with 20 mL of acetone was added dropwise into the flask for 1 h. After the addition was completed, and the solution continued to react for 6 h and was then filtered to collect the precipitate. The precipitate was washed with acetone twice and then dried under vacuum to obtain the hygroscopic yellow-white product. The yield was 85%. The product styrene-N,N-dimethylaminopropyl maleamidic acid (SDMAPMA) was subjected to elemental analysis. Elemental analysis for $C_{17}H_{24}O_3N_2$: Calcd: C, 67.10%; H, 7.89%; N, 9.21%. Found: C, 65.92%; H, 8.75%; N, 8.18%; amidoacidation degree, 88.8 mol %.

Quaternization of SDMAPMA

The above polymeric amidic acid, SDMAPMA (4.5 g), was dissolved in 80 mL of DMSO at 80°C and stirred until it was completely dissolved. When the temperature cooled down to 30°C, 2.5 g of MI mixed with 20 mL of DMSO was added dropwise for 1 h. After the addition was completed, the solution continued to react for 8 h, then was kept still for several hours. The solution was precipitated with tetrahydrofuran (THF), and the product was repeatedly dissolved and precipitated from methanol with THF, and then dried at 80°C for 24 h under vacuum to obtain the quaternary ammonium salt product with 80% yield. The product, orange-red hygroscopic MI quaternized SDMAPMA (MIQSDMAPMA), was obtained and analyzed by elemental analysis. Elemental analysis for $C_{18}H_{27}O_3N_2I$: Calcd: C, 48.43%; H, 6.05%; N, 6.27%. Found: C, 48.47%; H, 6.35%; N, 4.84%; degree of quaternization, 77.2%.

Viscometric Measurements

Viscometric measurements were performed with an Ubbelohde viscometer, which has a flow time of 72.89 s with deionized water ($18M\Omega$), at 30 ± 0.01°C. The polymer samples were dissolved in the salt solution and the polymer salt solution

was adjusted to yield a stock solution of 0.1 g/25 mL solvent. Viscosity data were calculated according to the Mark-Huggins equation:

$$\eta_{sp}/C = [\eta] + k'[\eta]^2 C \qquad (1)$$

where η_{sp} is the specific viscosity, C is the concentration of polymer, $[\eta]$ is the intrinsic viscosity, and k' is the Huggins constant.

Reduced Viscosity in Various pH Solutions

The method was the same as for the above-mentioned viscometric measurements. The pH values of the solutions were adjusted by adding HCl and NaOH in water.

RESULTS AND DISCUSSION

Characterization of SMA, SDMAPMA, and MIQSDMAPMA copolymers

SMA copolymer was confirmed by elemental analysis as an alternating copolymer.[38] The SMA copolymer with an M_w of 87,000 determined using the Mark-Houwink equation, $[\eta] = KM^\alpha$, where $K = 3.98 \times 10^{-4}$ and $\alpha = 0.596$,[39] at 25°C in THF solution (as described in Scheme 1){SCHEME 1} was then converted to SDMAPMA and MIQSDMAPMA. The characteristic absorption peaks of IR spectra are 1856, 1779, 1224 cm^{-1}; 3470, 1725, 1650, 1550 cm^{-1}; and 3470, 1725, 1650, 1550, 963 cm^{-1} for SMA, SDMAPMA, and MIQSDMAPMA, respectively.

Viscosity Measurement

Polyelectrolytes usually exhibit properties in solution that are quite different from general polymers when they are ionized in aqueous solution: the mutual repulsion of their charges causes the chain to expand. The size of the polyelectrolyte random coil is a function of the concentrations of the polymer and added salt, because both influence the degree of ionization. The property of viscosity is strongly affected by chain expansion. The reduced viscosity may increase as polymer concentration decreases, with a consequent increase in the degree of ionization of the polymer. However, the addition of low molecular weight electrolyte (salt) to the aqueous solution increases the ionic strength of the solution outside the polymer coil relative to the inside, and also reduces
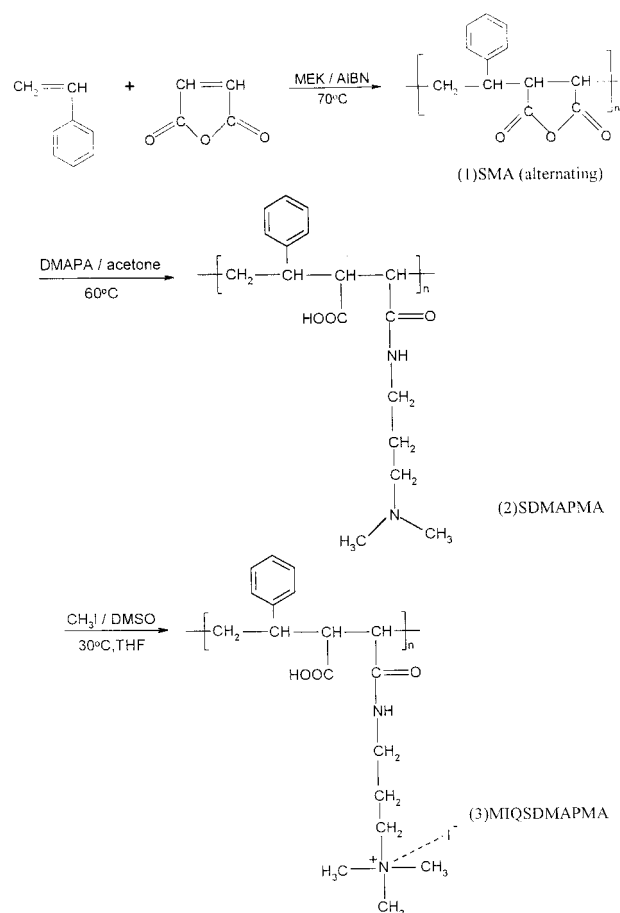
Scheme 1 Synthesis of SMA(1), SDMAPA(2), and MIQSDMAPMA(3) copolymer.

the number of site-bound counterions around the chain. Those effects cause the chain to contract.[40] In our experiments, the poly(MIQSDMAPMA) side chains would bear positive charges because the iodide ions became ionized in an aqueous solution. In this situation, the behavior of reduced viscosity exhibited a typical plot in pure water as shown in Figure 1(a). This result shows that the reduced viscosity in a higher concentration region sharply decreases with an increase in polymeric concentration because of the interaction of the polymer chain. After a maximum at the concentration of 0.1 g/dL is reached, the reduced viscosity then decreases. This phenomenon was also observed in MIQSDMAPM copolymer.[36] As shown by plotting of reciprocal reduced viscosity versus square root of polymeric concentration [Fig. 1(b)], the concentration dependence of the reduced viscosity conforms to the Fuoss equation,[41] $\eta_{sp}/C = A/(1 \times B\sqrt{C})$, where A corresponds to intrinsic viscosity $[\eta]$, B is a constant related to
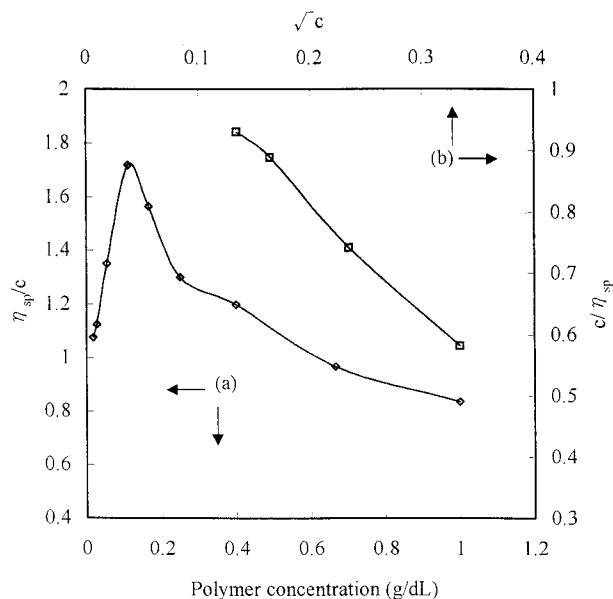
Figure 1 Reduced viscosity of poly(MIQSDMAPMA) in pure water. (a) Dependence of reduced viscosity on polymer concentration. (b) Dependence of reciprocal reduced viscosity on square root of polymer concentration.

an interaction of counterions with polyion, and C is polymeric concentration in the lower polymeric concentration region.

Effect of Various Salts on Intrinsic Viscosity of Poly(MIQSDMAPMA) in Aqueous Salt Solution

The addition of salt to the aqueous solution of polyelectrolyte causes the polymeric chain to contract and decrease the intrinsic viscosity, as mentioned above. The investigation of the influence of various salts on this effect and the interpretation of charged neutralization are presented in the following section.

Influence of Different Cations with a Common Anion (Cl⁻) on Intrinsic Viscosity of Poly(MIQSDMAPMA)

The intrinsic viscosity $[\eta]$ and Huggins constant k' can be calculated from Figures 2 and 3, and eq. (1). The data shown in Table I exhibit an increase in the intrinsic viscosity in the order of $Li^+ > Na^+ > K^+$ for LiCl, NaCl, and KCl, and in the order of $Ca^{2+} > Sr^{2+} > Ba^{2+}$ for $CaCl_2$, $SrCl_2$, and $BaCl_2$, respectively, for poly(MIQSDMAPMA) in $0.25M$ aqueous salt solution. These results follow the aqueous solution properties of polyelectrolytes
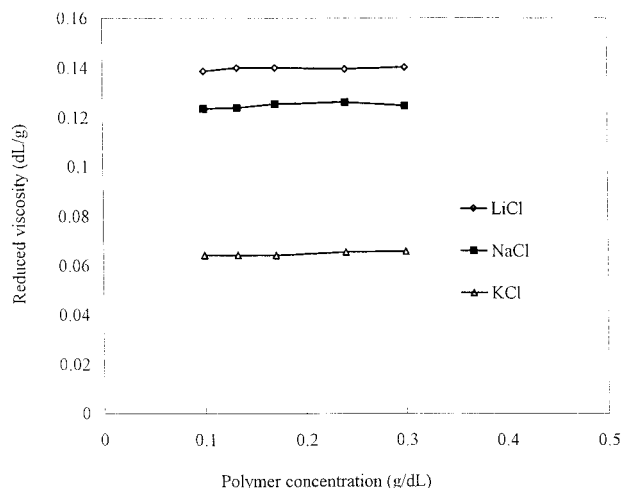

Figure 2 Reduced viscosity of poly(MIQSDMAPMA) versus polymer concentration in $0.25M$ aqueous solution of salts with monovalent cations.

such as poly(MIQSDMAPM)[36] and the Pearson principle,[42] but are contrary to polyampholyte behavior.[33,38,40]

Influence of Different Anions with a Common Cation (K⁺ or Na⁺) on Intrinsic Viscosity of Poly(MIQSDMAPMA)

For various halide anions, the data shown in Figure 4 and Table II indicate an increase of the intrinsic viscosity for monovalent anions in the order $F^- > Cl^- > Br^- > I^-$ (except for KI measured in $0.05M$) for KF, KCl, KBr, and KI, respec-
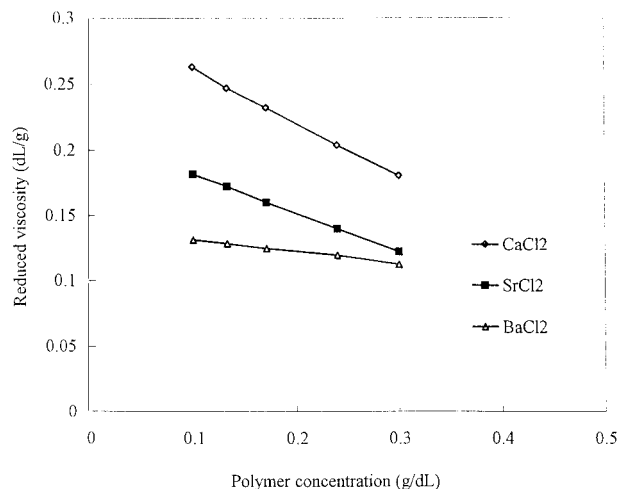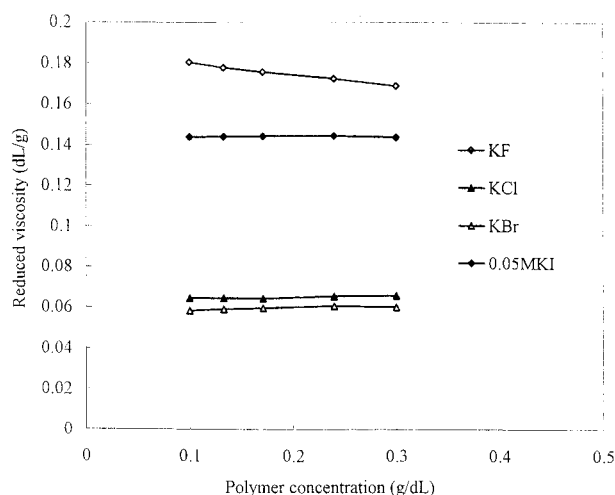

Figure 3 Reduced viscosity of poly(MIQSDMAPMA) versus polymer concentration in $0.25M$ aqueous solution of salts with various divalent cations.

Table I   Effect of Various Cations on the Viscosity Behavior of Poly(MIQSDMAPMA) at 30°C

Salt Solution (0.25M)	Slope	$[\eta]$	k'
LiCl	0.01	0.14	0.19
NaCl	0.01	0.12	0.45
KCl	0.01	0.06	2.00
CaCl$_2$	−0.41	0.30	−4.48
SrCl$_2$	−0.30	0.21	−6.67
BaCl$_2$	−0.09	0.14	−4.66

Table II   Effect of Various Halide Anions on the Viscosity Behavior of Poly(MIQSDMAPMA) at 30°C

Salt Solution (0.25M)	Slope	$[\eta]$	k'
KF	−0.06	0.19	−1.62
KCl	0.01	0.06	2.00
KBr	0.01	0.06	3.04
0.05MKI	0.00	0.14	0.02

tively, for poly(MIQSDMAPMA) in 0.25M aqueous salt solution. This is because the larger anion with common cation (K$^+$) (charge density is smaller) is easily polarized and bound to the quaternary ammonium group (R$_4$N$^+$) on poly(MIQSDMAPMA). The positive charge on polymeric side chains could therefore become effectively neutralized by the larger anion and reduce the intrinsic viscosity of poly(MIQSDMAPMA) in 0.25M aqueous salt solution. This result also conforms to the aqueous solution properties of poly(MIQSDMAPM)[36] and poly(MIQADMAPM).[37]

For various acidic groups, Figure 5 and Table III show an increase of the intrinsic viscosity for the present copolymer in 0.25M aqueous salt solution in the order ClO$_4^-$ > ClO$_3^-$; NO$_3^-$ > NO$_2^-$; HCO$_3^-$ > CH$_3$COO$^-$ for NaClO$_4$, NaClO$_3$, NaNO$_3$, NaNO$_2$, NaHCO$_3$, and CH$_3$COONa, respectively. These results are obviously contrary to our previ-

ous reports in cationic polyelectrolytes,[35–37] but conform to the behavior of polysulfobetaines such as ADMMAPS[44] in these aqueous salt solutions. This interesting phenomenon may be attributed to the presence of amidic acid group on the polymeric side chain during imidoacidation reaction.

For the divalent acidic groups, the data (Fig. 6 and Table IV) show an increase of the intrinsic viscosity for the present copolymer in 0.25M aqueous salt solution in the order CO$_3^{-2}$ > S$_2$O$_3^{-2}$ (0.01M) > SO$_3^{-2}$ > SO$_4^{-2}$ for Na$_2$CO$_3$, Na$_2$SO$_3$, Na$_2$SO$_4$, and Na$_2$S$_2$O$_3$, respectively. The poly(MIQSDMAPMA) does not dissolve in 0.25M aqueous solution of KI, NaNO$_3$, and Na$_2$S$_2$O$_3$. This phenomenon is attributed to the attraction of the polymeric side chains by salt counterions (I$^-$, NO$_3^-$, and S$_2$O$_3^{-2}$) and this attraction prevents the polymeric chain from expanding and results in agglomerating. This tendency obeys the Pearson principle and is similar to the previous
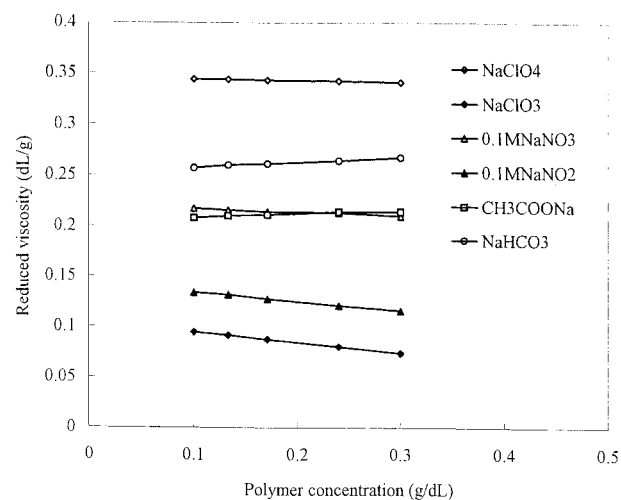

Figure 4   Reduced viscosity of poly(MIQSDMAPMA) versus polymer concentration in 0.25M aqueous solution of salts with various halide anions.

Figure 5   Reduced viscosity of poly(MIQSDMAPMA) versus polymer concentration in 0.25M aqueous solution of salts containing various monovalent acid groups.
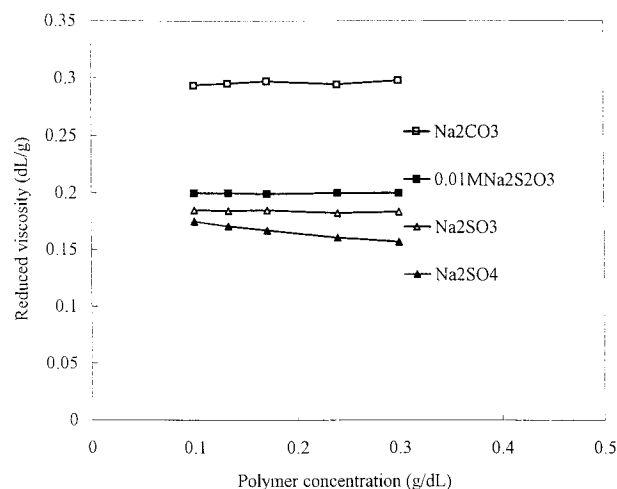
Table III   Effect of Various Monovalent Acid Groups on the Viscosity Behavior of Poly(MIQSDMAPMA) at 30°C

Salt Solution (0.25M)	Slope	$[\eta]$	k'
NaClO$_4$	-0.02	0.35	-0.14
NaClO$_3$	-0.11	0.11	-9.53
0.1M NaNO$_3$	-0.04	0.22	-0.71
0.1M NaNO$_2$	-0.09	0.14	-4.49
NaHCO$_3$	0.05	0.25	0.76
CH$_3$COONa	0.03	0.21	0.71

Table IV   Effect of Various Divalent Acid Groups on the Viscosity Behavior of Poly(MIQSDMAPMA) at 30°C

Salt Solution (0.25M)	Slope	$[\eta]$	k'
0.01M Na$_2$S$_2$O$_3$	0.00	0.20	0.02
Na$_2$SO$_4$	-0.09	0.18	-2.70
Na$_2$SO$_3$	-0.01	0.19	-0.27
Na$_2$CO$_3$	0.01	0.29	0.14

results of poly(TMAAI), poly (MIQSDMAPM), and poly(MIQADMAPM).[35–37]

Effect of Various Salt Concentrations in Aqueous Solution on the Intrinsic Viscosity of Poly(MIQSDMAPMA)

The presumed concept of chain contraction of the cationic polymers for salt ions attracting or site-binding on the polymers was confirmed. Therefore, the phenomenon of a decrease in the reduced viscosity with an increase of salt concentration was rationalized. The effect of various NaCl concentrations between 0.05 to 1.0M on the reduced viscosity is illustrated in Figure 7 for the poly(MIQSDMAPMA). A significant decrease in the reduced viscosity apparently occurred with an increase of the NaCl concentration. These phenomena are similar to the poly(MIQSDMAPM),[36]

but in sharp contrast with the inner salt of poly-sulfobetaines.[33,38,43]

When the salt concentration is increased, some negative charges of salt would become site-bound on the quaternary ammonium group (R$_4$N$^+$) on the poly(MIQSDMAPMA). The polymeric charges would become neutralized by the counterions in the aqueous solution. This occurrence would reduce the repulsion of polymeric side chains resulting in both decreasing the extent of tightly stretched chains and coiling the polymeric main chains. The chloride ions (Cl$^-$) binding on the quaternary ammonium group (R$_4$N$^+$) of poly(MIQSDMAPMA) were increased in high salt concentration to form regions of high local charge density. Above the saturated binding concentration, the strong interaction between salt and polymer pulls the distance between polymer and polymer closer to make polymer side chains twisted around each other. This action makes polymer main chains wind around polymer side chains and agglomerate. At this moment, Huggins con-
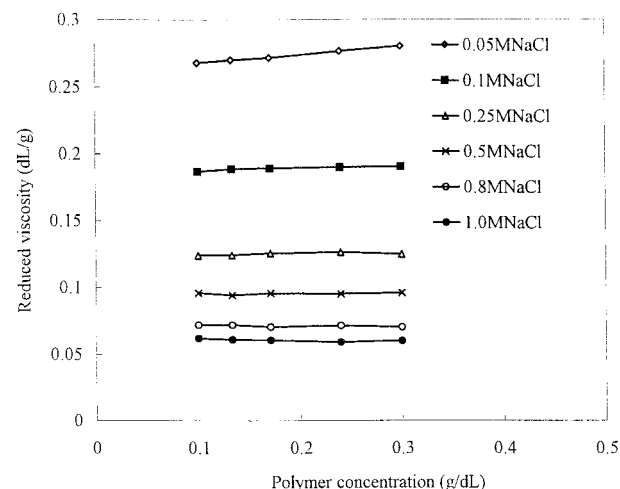

Figure 6   Reduced viscosity of poly(MIQSDMAPMA) versus polymer concentration in 0.25M aqueous solution of salts containing various divalent acid groups.

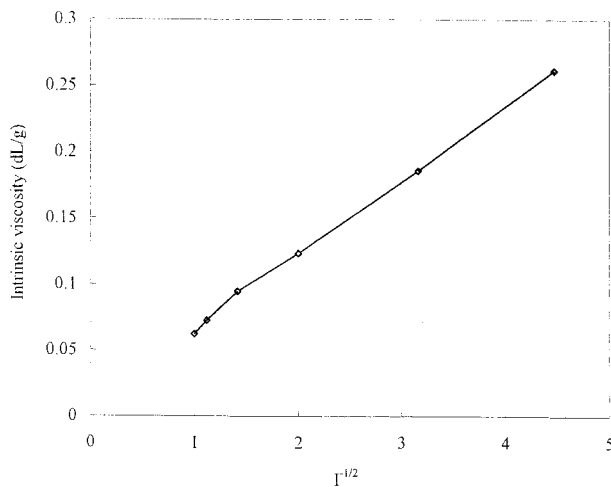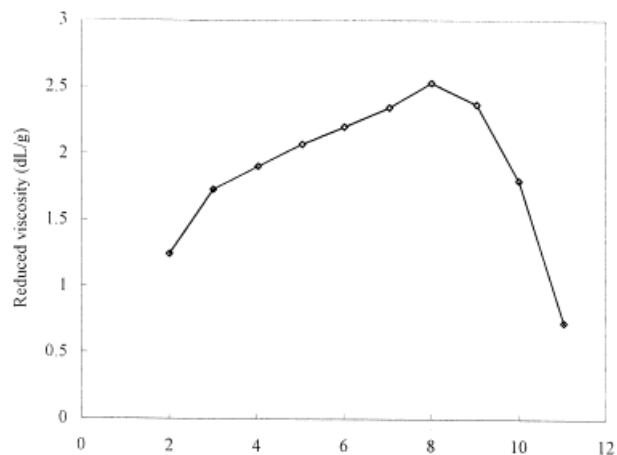**Figure 7**   Reduced viscosity of poly(MIQSDMAPMA) as a function of various concentrations of NaCl.

Table V   Effect of NaCl Concentrations or Ionic Strength on the Intrinsic Viscosity and Huggins Constant of Poly(MIQSDMAPMA) at 30°C

NaCl (M)	$I^{-1/2}$ (Ion Strength)	Slope	$[\eta]$	k'
0.05	4.472	0.06	0.26	0.91
0.10	3.162	0.02	0.19	0.49
0.25	2.000	0.01	0.12	0.45
0.50	1.414	0.00	0.10	0.22
0.80	1.118	−0.01	0.07	−1.39
1.00	1.000	−0.01	0.06	−2.68

Figure 9   A plot of reduced viscosity versus pH for poly(MIQSDMAPMA) in 0.2 g/dL aqueous solution.

stant k' decreases to the smallest. This situation therefore coincides with that of the poly(MIQSD-MAPMA) which does not dissolve in 0.25M KI, NaNO₃, and Na₂S₂O₃ aqueous solution.

The slope, intrinsic viscosity, and Huggins constant k' shown in Table V can be calculated through Figure 7 and eq. (1). The slopes decrease with the increase in the concentration of simple electrolyte (NaCl) and change from positive value to negative value. The values for Huggins constants and the intrinsic viscosities also decrease with increase in the concentration of the salt.

The above phenomena might be attributed to the addition of mobile simple electrolyte which can loosen the compact structure that results from the inter- and intramolecular ionic interactions to reduce electrical repulsive force from polymer chains, thus enabling the polymer to behave more freely in the salt solution or recoil to agglomerate.[16,36] In other words, the higher the

salt concentration, the lower the electrostatic repulsive force and, thus, the lower the intrinsic viscosity. This result was also observed by Liaw et al.,[16] Lee et al.,[35–38,43,44] Conway,[45] and Eisenberg and Pouyet.[11] The intrinsic viscosity $[\eta]$, obtained by extrapolating the curves in Figure 7 at constant simple electrolyte concentration to zero concentration of the polymer, is plotted in Figure 8 as a function of the reciprocal square root of the ionic strength ($I^{-1/2}$). A linear relationship obtained from Figure 8 conforms to the general law of polyelectrolyte.[46,47]

Effect of Various pH Aqueous Solutions on the Reduced Viscosity of Poly(MIQSDMAPMA)

To investigate the effect of pH values on the reduced viscosity of the present copolymer, poly(MIQSDMAPMA), the copolymer was dissolved in various pH solutions which were adjusted by adding HCl or NaOH. The results shown in Figure 9 indicate that there is a maximum reduced viscosity at pH 8. This result implies that the maximum net charge repulsion occurs at this pH condition for the present copolymer.

CONCLUSION

The aqueous solution properties of cationic polyelectrolyte were evidently changed as salts were added to polymeric aqueous solution. As the radius of the salt ion was increased, the degree of attraction for poly(MIQSDMAPMA) and salt, and

Figure 8   Intrinsic viscosity of poly(MIQSDMAPMA) as a function of reciprocal square root of ionic strength.

the degree of polymeric recoiling, increased. Simultaneously, the reduced viscosity and intrinsic viscosity decreased. As the salt concentration was increased, the intrinsic viscosity decreased and Huggins constant k' decreased (Huggins constant k' was influenced by polymer–solvent and polymer–polymer interaction). Above the saturated binding concentration of NaCl aqueous solution, the strong interaction between salt and polymer make polymer main chains wind around polymer side chains and agglomerate. This situation therefore coincides with that of poly(MIQSD-MAPMA) which does not dissolve in $0.25M$ KI, $NaNO_3$ and $Na_2S_2O_3$, aqueous solution.

Although the tendency of intrinsic viscosity for MIQSDMAPMA in various aqueous salt solutions is similar to that of other cationic polyelectrolytes exclusive of monovalent acid group salts, the Huggins constant for MIQSDMAPMA shows an extreme distinction from MIQSDMAPM, MIQAD-MAPM copolymer, or poly(TMAAI) homopolymer. This phenomenon is mainly attributed to the presence of the styrene segments and carboxylic groups, hydrophobic/hydrophilic groups, in the pendent chains of the MIQSDMAPMA copolymer. The reduced viscosity for the present copolymer in various pH solutions showed a maximum value at pH 8.

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