Poly(sulfobetaine)s and Corresponding Cationic Polymers. IX. Synthesis and Aqueous Solution Properties of Zwitterionic Poly(sulfobetaine) Derived from a Styrene–*N,N*-Dimethylaminopropyl Maleamidic Acid Copolymer

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ABSTRACT: A styrene–*N,N*-dimethyl(maleamidic acid)-propyl ammonium propane sulfonate (SDMMAAPS) copolymer was synthesized through an amidoacidation reaction of a styrene–maleic anhydride alternating copolymer with *N,N*-dimethylaminopropylamine (ring-opening reaction) and then reacted with propane sultone. The cloud point and minimum salt concentration (msc) of this ampholytic SDMMAAPS copolymer were determined in aqueous salt solutions. The effects of counterions on the cloud point and msc of SDMMAAPS were not entirely the same as those of other zwitterionic poly(sulfobetaine)s. The greatest difference from other poly(sulfobetaine)s, such as styrene–*N,N*-dimethyl(maleimido propyl)ammonium propane sulfonate copolymers, was the carboxylic group on the polymer chain unit of SDMMAAPS. © 2003 Wiley Periodicals, Inc. J Appl Polym Sci 89: 1884–1889, 2003

INTRODUCTION

Synthetic polyampholytes can be prepared from zwitterionic monomers, which exhibit the requisite net charge of zero at the appropriate pH. A quaternary ammonium group provides the positive charge, and the negative charge is provided by a carboxylate or sulfonate group (carboxybetaine or sulfobetaine). A number of investigations have focused on the unusual properties of zwitterionic polyampholytes.

Zwitterionic monomers of sulfobetaines or carboxybetaines derived from dimethylamino alkylacrylates and dimethylamino acrylamides have widely been used in industries manufacturing textiles, medical products, charged dispersing agents, protective colloids, and related materials.[1–9] The synthesis and aqueous solution properties of these betaine monomers have been reported by several researchers.[10–25]

During studies[26–31] of the binding interactions of salt ions and polymers with respect to the Huggins constant, *k'*, the counterion size was found to affect the degree of binding for salt-ion-attracting polymers;[12–16,19,22–24,32] that is, soft salt anions and cations were more effective solubilizers than hard anions and cations.[33,34]

A series of poly(sulfobetaine)s and corresponding cationic polymers prepared from zwitterionic sulfobetaine and corresponding cationic monomers with different electron-withdrawing groups and with variations in the methylene units between the charge groups were previously researched in our laboratory. The solution properties of poly[*N,N'*-dimethyl(acrylamidopropyl)ammonium propane sulfonate] [poly(D-MAAPS)] and poly[*N,N'*-dimethyl(methacryloyloxyethyl)ammonium propane sulfonate] [poly(DMAPS)] were reported in previous articles.[22,25,34,35] In the presence of salts or electrolytes in aqueous solutions, because of the inter or intra dipolar ionic interaction, the degree of polymeric chain extension or recoiling changes the viscosity. The previous poly(DMAAPS) article concluded that soft salt anions and cations bound easily to the quaternary ammonium and sulfonate groups of poly(DMAAPS).[25] Recently, we have concentrated on polyampholytes prepared from copolymers having high purities and similar structural characteristics. For example, styrene–*N,N*-dimethyl-(maleimido propyl)ammonium propane sulfonate (SDMMAPS) and acrylamide–*N,N*-dimethyl maleimido propyl ammonium propane sulfonate (ADMMAPS) copolymers were prepared from a styrene–maleic anhydride (SMA) copolymer and an acrylamide–maleic anhydride copolymer, respectively. The properties of these ampholytic SDMMAPS and ADMMAPS copolymers in aqueous salt solutions were studied by the measurement of the cloud point and intrinsic viscosity.[36,37] Even though the tendency of the minimum salt concentration (msc) and intrinsic viscosity for poly[styrene–*N,N*-dimethyl(maleimido propyl)ammonium propane sulfonate] [poly(SDMMAPS)] in various aqueous salt solutions was similar to that of other zwitterionic poly(sulfobetaine)s, the

Huggins constant for poly(SDMMAPS) in aqueous salt solutions showed an extreme distinction from that of sulfobetaine homopolymers, such as [poly(D-MAAPS], or copolymers, such as poly(acrylamide–*N*,*N*-dimethyl maleimido propyl ammonium propane sulfonate) [poly(ADMMAPS)].[34,35,37] This phenomenon can be attributed mainly to the presence of the styrene (ST) segment, a hydrophobic group, pendent to the SDMMAPS copolymer.

The synthesis and properties of poly[styrene–*N*,*N*-dimethyl(maleamidic acid) propyl ammonium propane sulfonate] are not discussed in existing related reports. They can be compared with other polyampholytes with only betaine groups. The investigation of the cloud-point determination or msc for the copolymer in the presence of various salts is the main focus of this article.

EXPERIMENTAL

Materials

ST, *N*,*N*-dimethylaminopropyl amine (DMAPA), dimethyl sulfoxide (DMSO), methyl ethyl ketone (MEK), and propane sultone (PS) were used as received. Maleic anhydride (MA; mp = 56°C) and azobisisobutyronitrile (AIBN) were purified by recrystallization in methanol. Methanol and acetone were distilled twice. Deionized water had a resistivity of 18 MΩ.

Preparation of the SMA copolymer

ST (18.237 g, 0.175 mol), MA (17.162 g, 0.175 mol), and MEK (150 mL) were introduced into a 0.25-L flask equipped with a stirrer, a cooler, and a thermometer. The contents were stirred at 80°C for complete dissolution. To the solution, AIBN (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 a nitrogen atmosphere. The solution was then precipitated with diethyl ether. The product was repeatedly dissolved and precipitated from acetone with diethyl ether and was then dried at 80°C for 24 h under vacuum, yielding the SMA copolymer. The yield was 98%. The polymer was analyzed by elemental analysis.

ELEM. ANAL. Calcd. for $C_{12}H_{10}O_3$ [weight-average molecular weight (M_w) = 202]: C, 71.28%; H, 4.98%; O, 23.74%. Found: C, 70.22%; H, 5.61%; O, 24.12%.

Amidoacidation of the SMA copolymer for the preparation of styrene–*N*,*N*-dimethylaminopropyl maleamidic acid (SDMAPMA)

The SMA copolymer (10.6 g) was mixed with 200 mL of acetone in a 500-mL, single-necked flask at 55°C and was stirred until it was completely dissolved. When the temperature cooled down to 40°C, 6.75 g of 3-di-

methylaminopropylamine, mixed with 20 mL of acetone, was added dropwise to the flask for 1 h. After the addition was completed, the solution continued to react for 6 h, and then it was filtered foer the collection of the precipitate. The precipitate was washed with acetone twice and then dried under vacuum; a hygroscopic, yellow-white product was obtained. The yield was 85%. The product SDMAPMA was analyzed by elemental analysis.

ELEM. ANAL. Calcd. for ($C_{17}H_{24}O_3N_2$): C, 67.10%; H, 7.89%; N, 9.21%. Found: C, 65.92%; H, 8.75%; N, 8.18%. Amidoacidation degree: 88.8%.

Preparation of poly(SDMMAAPS)

The aforementioned 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 50°C, 2.3 g of PS, mixed with 30 mL of DMSO, was added dropwise for 1 h. After the addition was completed, the solution continued to react for 8 h and then was allowed to stand overnight. The precipitated viscous product was collected by filtration, washed with methanol for the removal of PS, and dried under reduced pressure for 24 h; the styrene–*N*,*N*-dimethyl(maleamidic acid)propyl ammonium propane sulfonate (SDMMAAPS) copolymer product in an 80% yield was obtained. Orange-yellow, hygroscopic poly(SDMMAAPS) was obtained and analyzed by elemental analysis.

ELEM. ANAL. Calcd. for $C_{20}H_{30}O_6SN_2$ (M_w = 426): C, 56.33%; H, 7.04%; N, 6.57%. Found: C, 57.64%; H, 7.42%; N, 5.78%. Degree of quaternization: 87.9%

Determination of the cloud point

Each SDMMAAPS copolymer (0.1 g) was dissolved in an appropriate aqueous salt solution (10 mL). Cloud-point titration was used to determine the point at which the polymer became insoluble in an aqueous salt solution (25 ± 5 mL); it was performed by the dissolution of the material in an aqueous salt solution with sufficient salt content and by the titration of the stirred solution with deionized water to the first indication of turbidity. The salt concentration approaching the cloud point was the msc.

RESULTS AND DISCUSSION

Characterization of the SMA, SDMAPMA, and SDMMAAPS copolymers

The SMA copolymer was confirmed by elemental analysis as an alternating copolymer.[36] The SMA copolymer had an M_w value of 87,000, as determined with the Mark–Houwink equation, $[\eta] = K\bar{M}^\alpha$, where $[\eta]$ is the intrinsic viscosity, $\bar{M}$ is the average molecular weight, K is 3.98×10^{-4}, and α is 0.596,[38] at 25°C in
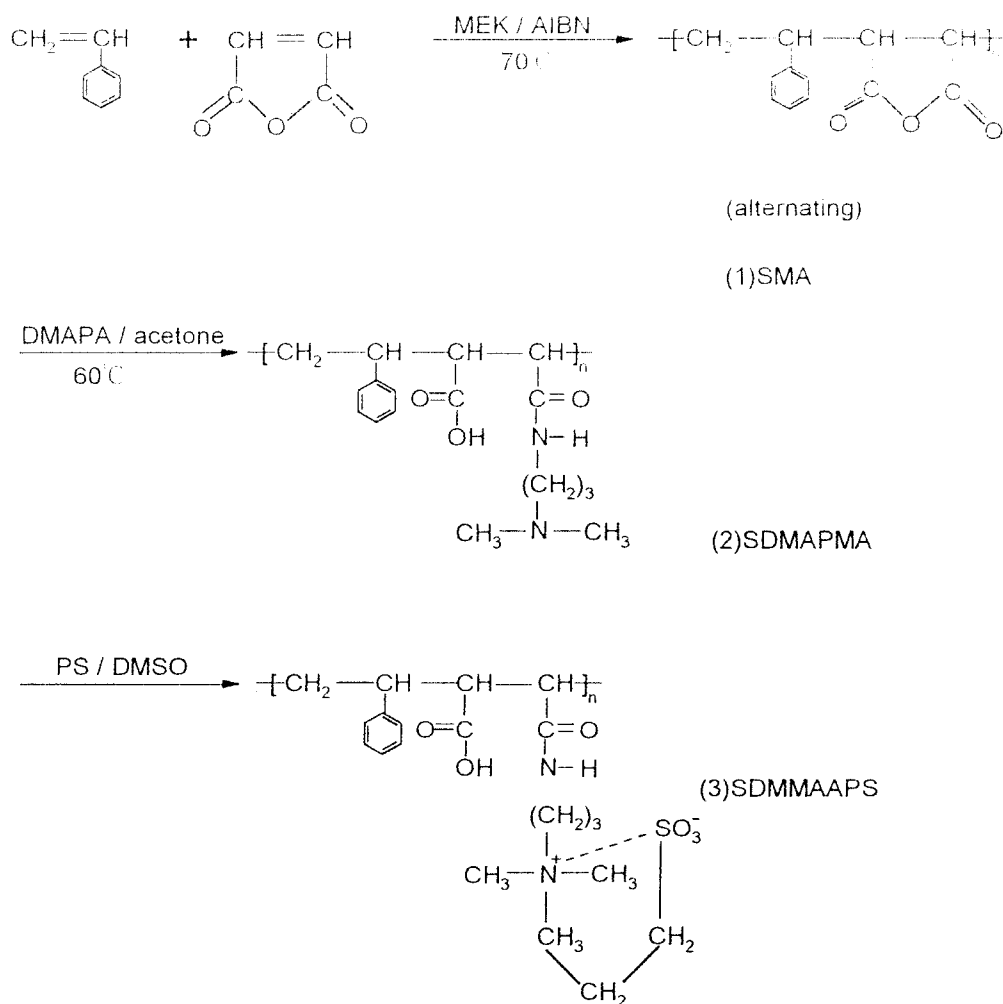
Scheme 1 Synthesis of (1) SMA, (2) SDMAPMA, and (3) SDMMAAPS copolymers.

a tetrahydrofuran solution. It was then converted into SDMAPMA and SDMMAAPS (as described in Scheme 1). The characteristic absorption peaks of the IR spectra were 1856, 1779, 1224, and 800–600 for SMA, 3600–2400, 1725, and 1640–1550 for SD-MAPMA, and 1183 and 1042 cm^{-1} for SDMMAAPS.

Determination of the cloud point

The poly(SDMMAAPS) copolymers were insoluble in water but formed transparent gels in aqueous solutions. The positive charges on the poly(SDMMAAPS) units were attracted to the negative charge sites, forming an ionically crosslinked network. For this reason, salts were added to the polymeric aqueous solutions to disrupt the ionic interactions and dissolve the polymers.

In the aqueous salt solutions of poly(SDMMAAPS) copolymers, the cations (or anions) of the salts were site-bound on the sulfonate group [SO_3^-; or the quaternary ammonium group (R_4N^+)] of poly(SDM-MAAPS). By collapsing the ionically crosslinked network and expanding the polymeric chain, poly(SDM-

MAAPS) dissolved in aqueous salt solutions.[25,34–37] To study the solution behavior of poly(SDMMAAPS), we needed to measure the msc required for solubilization of the polymer.

Effect of different cations with a common anion (Cl$^-$) on the msc of poly(SDMMAAPS)

Poly(SDMMAAPS) has a —COOH group and a sulfobetaine group on the side chain. Because these two groups cause a contrary effect on the cloud point of poly(SDMMAAPS) in aqueous salt solutions, to determine the cloud point, which is the msc required for poly(SDMMAAPS) to be soluble in an aqueous salt solution, we must consider these two effects to draw a balance. That is, salt ions attract poly(SDMMAAPS) to open the inner salt ring of polyampholyte and, at the same time, complex with —COO$^-$ or abstract H$^+$ from —COOH side groups to reduce the degree of site binding on sulfobetaine groups; therefore, the msc of poly(SDMMAAPS) in aqueous salt solutions differs from that of general poly(sulfobetaine)s containing just a single sulfobetaine group,[25,34–37] such as poly-

TABLE I
MSC of the SDMMAAPS Copolymer in Salt Solutions
for Various Cations with a Common Anion at 30°C

Salt	MSC $(M) \times 10^2$
LiCl	8.5
NaCl	9.3
KCl	10.3
CaCl$_2$	3.3
SrCl$_2$	3.4
BaCl$_2$	3.6

(DMAAPS), poly(SDMMAPS), and poly(ADMMAPS). The data in Table I show that the msc of poly(SDM-MAAPS) in aqueous salt solutions decreased in the order $Li^+ < Na^+ < K^+$ and $Ca^{2+} < Sr^{2+} < Ba^{2+}$ for LiCl, NaCl, KCl, and CaCl$_2$, SrCl$_2$, and BaCl$_2$, respectively. These results are contrary to conventional poly(sulfobetaine)s. The reason is that —COO$^-$ from —COOH side groups of poly(SDMMAAPS) is a hard base preferring to bind with a hard acid that has a small radius, such as Li^+ or Ca^{2+}. The smaller the radius is of a cation, the stronger the binding ability is on —COOH to make the carboxylic (—COOH) side group easily transform into a carboxylate (—COO$^-$) anionic polyelectrolyte. This effect is greater than that when the ions dissociate from the salts to site-bind on the sulfonate group [SO$_3^-$; or the quaternary ammonium group (R$_4$N$^+$)] of poly(SDMMAAPS); that is, the polyelectrolyte effect for Li^+ or Ca^{2+} is greater than the polyampholyte effect [see Scheme 2(a)].

Effect of different halide anions with a common cation (K$^+$) on the msc of poly(SDMMAAPS)

The cloud points of the copolymer solutions containing salts with a common cation (K$^+$) but different halide anions are shown in Table II. For these salts, the data indicate the increased solubility of the copolymer in the order $Cl^- < Br^- < I^-$ for KCl, KBr, and KI, respectively. The reason is that the ability of the halide anion abstracting H$^+$ from —COOH side groups to make the —COOH side group transform into a —COO$^-$ anionic polyelectrolyte decreases in the order $Cl^- > Br^- > I^-$ for KCl, KBr, and KI, respectively, and this effect is smaller when the ability of halide anion site binding on the quaternary ammonium group (R$_4$N$^+$) of sulfobetaine to open the inner ring of poly(SDMMAAPS) is greater. Therefore, after these two effects compete with each other, the effect of the ability of the halide anion to open the inner ring of sulfobetaine is predominant in deciding the solubility trend of poly(SDMMAAPS) in aqueous salt solutions; that is, the polyampholyte effect for Cl^-, Br^-, and I^- is greater than the polyelectrolyte effect under this condition.

However, the cloud point of the fluoride anion (F$^-$) has not been found because the msc needed for the solubilization of poly(SDMMAAPS) in KF$_{(aq)}$ is very small and the F$^-$ ion belongs to a strong hard base. This phenomenon is attributed to the very small radius of the F$^-$ anion resisting polarization; the F$^-$ anion has no capability for site binding on the sulfobetaine group to open the inner salt ring of poly(SDM-MAAPS) but strongly attracts H$^+$ from —COOH side groups to make the —COOH side group totally transform into a —COO$^-$ anionic polyelectrolyte, which produces an electrostatic repulsive force leading to the full expansion of the polymer chain in aqueous salt solutions. This polyelectrolyte effect is predominant in making the F$^-$ anion become a much better solubilizer than other halide anions for the solubilization of poly(SDMMAAPS) in aqueous salt solutions [Scheme 2(b)].

Effect of different acid groups with a common cation (Na$^+$) on the msc of poly(SDMMAAPS)

The cloud points of the copolymer solutions containing salts with a common cation (Na$^+$) but different acid groups are shown in Table III. For the salts containing various monovalent acid groups, from our previous report for poly(methyl ioxide quarternized styrene-N,N-dimethylaminopropyl maleamidic acid) (poly(MIQSDMAPMA)),[39] we know that the various monovalent acid groups possess very small charge densities and are easily polarized to site-bind on the quaternary ammonium group (R$_4$N$^+$) on the side chains of poly(SDMMAAPS) and, at the same time, pull much closer to other side-chain groups of R$_4$N$^+$ and COOH between interpolymer and intrapolymer chains of poly(SDMMAAPS). This action lets the copolymer behave like an amino acid structure, which can bind together, producing a new ring of NR$_4^+$–COO$^-$, which is a polyampholyte structure, and it exhibits polycarboxybetaine behavior. At the same time, the copolymer changes into another strong anionic sulfonate (SO$_3$H) polyelectrolyte [as shown in Scheme 2(c)]. Therefore, the data in Table III indicate that no cloud points were found for the copolymer in various monovalent acid salts, except for sodium chlorate and sodium nitrate.

The increased solubility of the copolymer was in the order $NO_3^- > ClO_3^-$ for NaNO$_3$ and NaClO$_3$, respectively. This is the reason that no structure transformation happens for the copolymer in NaNO$_3$ and NaClO$_3$ and that the effect of attracting H$^+$, making carboxylic side groups dissociate into anionic carboxylate polyelectrolytes, is predominant in contrast to that of site binding on sulfobetaine groups to open the inner salt ring of poly(SDMMAAPS). However, poly(SDMMAAPS) exhibited a strong site-binding effect behavior, and no cloud point was found in the perchlorate solution. Because of dissociation into H$^+$ and CO$_3^{-2}$, HCO$_3^-$ loses the effect of making carboxylic acid side groups dissociate into anionic carboxylate polyelectrolytes and keeps the effect of
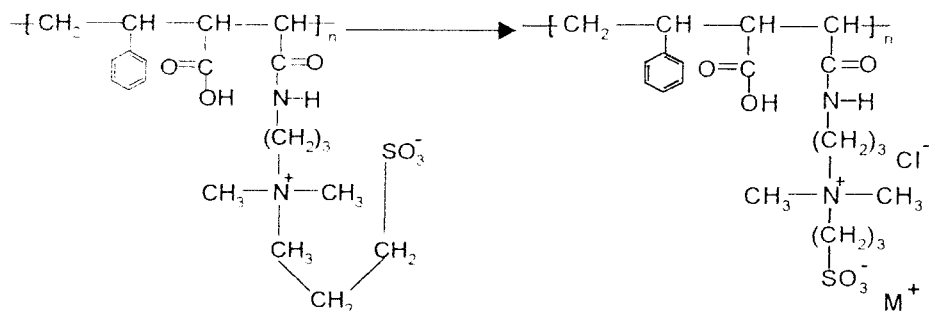
(a)
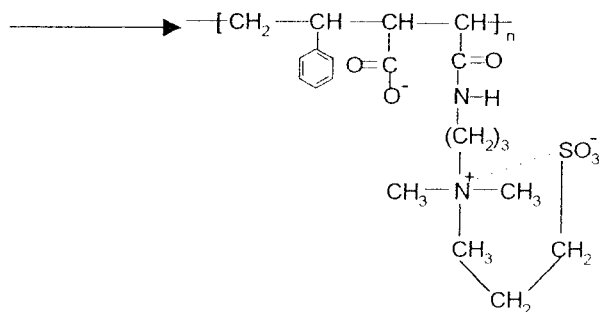
(b)
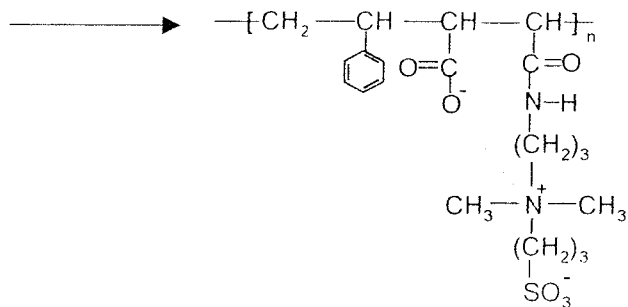
(c)

Scheme 2 Structural transformation of poly(SDMMAAPS) in various salt solutions: (a) the site-binding effect is greater than the H$^+$ abstracting effect and a cloud point is shown, (b) the H$^+$ abstracting effect is much greater than the site-binding effect and no cloud point is shown, and (c) the two effects are comparable and a cloud point is shown.

site binding on sulfobetaine groups to open the inner salt ring of poly(SDMMAAPS), which becomes predominant; therefore, HCO3$^-$ also has no cloud point.

TABLE II
MSC of the SDMMAAPS Copolymer in Salt Solutions for Various Halide Anions with a Common Cation at 30°C

Salt	MSC (M) $\times 10^2$
KF	< 0.1
KCl	10.3
KBr	9.6
KI	7.3

Similarly, the data in Table III show that cloud points were not observed in the salts containing various divalent acid groups, except for sodium sulfate. This was due to SO_4^{2-} attracting sulfobetaine groups to open the inner salt ring of poly(SDMMAAPS) and simultaneously abstracting H$^+$ from —COOH side groups to reduce the degree of salt-attracted site binding on sulfobetaine groups. Therefore, the msc of poly(SDMMAAPS) in $SO_{4(aq)}^{2-}$ was smaller than that of NO$_3^-$ and ClO$_3^-$ in aqueous salt solutions. The effect of salt abstracting H$^+$ from —COOH side groups plays a more important role in the cloud-point determination than the effect of site binding to sulfobetaine groups to open the inner salt.

TABLE III
MSC of the SDMMAAPS Copolymer in Salt Solutions for Various Acid Groups with a Common Cation at 30°C

Salt	MSC (M) $\times 10^2$
$NaClO_4$	< 0.1
$NaClO_3$	9.9
CH_3COONa	< 0.1
$NaHCO_3$	< 0.1
$NaNO_3$	8.9
$NaNO_2$	< 0.1
$Na_2S_2O_3$	< 0.1
Na_2SO_4	3.5
Na_2SO_3	< 0.1
Na_2CO_3	< 0.1

The msc in various aqueous salt solutions for SDM-MAAPS is the same as those of poly(DMAPS)[25] and poly(DMAAPS)[34,35] but is lower than that of SDM-MAPS[38] by one order of magnitude. This is due to the balance between the hydrophobic nature of the ST unit and the hydrophilic carboxylic side group (—COOH) unit in the copolymer, which keeps the solubility of poly(SDMMAAPS) the same as that of normal poly(sulfobetaine)s in salt solutions.

CONCLUSIONS

The behavior of poly(SDMMAAPS) in aqueous salt solutions is different from that of other poly(sulfobetaine)s in some respects. Poly(SDMMAAPS) possesses a —COOH group and a sulfobetaine group on the side chain. These two groups cause a contrary effect on the determination of the cloud point and msc of poly(S-DMMAAPS) in aqueous salt solutions. The experiments have shown that if the effect of abstracting H^+ from —COOH side groups for a salt anion is greater than that of site binding on sulfobetaine groups to open the inner salt ring of poly(SDMMAAPS), the cloud point is not observed. That is, the polymer readily dissolves in the salt solution. However, if the effect of site binding on the sulfobetaine group for a salt ion is predominant, in contrast to that of abstracting H^+ from —COOH side groups, the polymer aqueous salt solution shows a cloud point. The msc in various aqueous salt solutions for poly(SDMMAAPS) is the same as those of poly(DMAPS) and poly(D-MAAPS) but is lower than that of SDMMAPS by one order of magnitude.

References

1. Rohm and Hass Co. Brit. Pat. 1,077,772 (1967).
2. Bahr, U.; Wieden, H.; Rinkler, H. A.; Nischk, G. E. Makromol Chem 1972, 1, 161.
3. (a) Szita, J.; Bahr, U.; Wieden, H.; Marzolph, H.; Nischk, G. E. Belg. Pat. 659,316 (1965); (b) Szita, J.; Bahr, U.; Wieden, H.; Marzolph, H.; Nischk, G. E. Brit. Pat. (Amended) 1,024,029 (1966).
4. Spriestersbach, D. R.; Clarke, R. A.; Couper, M.; Patterson, H. T. U.S. Pat. 3,473,998 (1969).
5. (a) Ishikura, S.; Mizuguchi, R.; Takahashi, A. Jpn. Kokai Tokkyo Koho 80,386 (1977); (b) Ishikura, S.; Mizuguchi, R.; Takahashi, A. Jpn. Kokai Tokkyo Koho 80,387 (1977).
6. Thomson, E.; Parks, E.; Allan, K. Eur. Pat. Appl. 209,337 (1987).
7. Hagen, J.; Bischoff, M.; Heing, W. Eur. Pat. Appl. 72,509 (1983).
8. Ohme, R.; Rusche, R. J.; Seibt, J. H. Eur. Pat. Appl. 205,625 (1986).
9. Seibt, H.; Ballschuk, D.; Ohme, R.; Zastrow, L. Ger. Pat. 286,178 (1991).
10. Ladenheim, H.; Morawetz, H. J Polym Sci 1957, 26, 251.
11. Hart, R.; Timmerman, D. J Polym Sci 1958, 28, 638.
12. Salamone, J. C.; Volkson, W.; Raia, S. C. D. C.; Broggi, A.; Hsu, T. D. Polym Prepr 1973, 14, 731.
13. Salamone, J. C.; Volkson, W.; Israel, S. C.; Olson, A. P.; Raia, D. C. Polymer 1977, 18, 1058.
14. Salamone, J. C.; Volkson, W.; Olson, A. P.; Israel, S. C. Polymer 1978, 19, 1157.
15. (a) Monroy Soto, V. M.; Galin, J. C. Polymer 1984, 25, 121; (b) Monroy Soto, V. M.; Galin, J. C. Polymer 1984, 25, 254.
16. Galin, M.; Marchal, E.; Mathis, A.; Meurer, B.; Monroy Soto, V. M.; Galin, J. C. Polymer 1987, 28, 1937.
17. Schulz, D. N.; Kitano, K.; Danik, J. A.; Kaladas, J. J. In Polymer in Aqueous Media; Glass, J. E., Ed.; Advances in Chemistry Series 223; American Chemical Society: Washington, DC, 1989; p 165.
18. Asonova, T. A.; Razvodovskii, Y. F.; Zezin, A. B.; Kargin, V. A. Dokl Akad Nauk 1969, 118, 583.
19. Asonova, T. A.; Razvodovskii, Y. F.; Zezin, A. B. Vysokomol Soyed A 1974, 16, 777.
20. Topchiev, D. A.; Mkrtchyan, L. A.; Simonyan, R. A.; Lachinov, M. B.; Kabanov, R. A. Vysokomol Soyed A 1977, 19, 506.
21. (a) Laschewsky, A.; Zerbe, I. Polymer 1991, 32, 2070; (b) Laschewsky, A.; Zerbe, I. Polymer 1991, 32, 2081.
22. Liaw, D. J.; Lee, W. F.; Whung, Y. C.; Lin, M. C. J Appl Polym Sci 1987, 34, 999.
23. Itoh, Y.; Abe, K.; Senoh, S. Makromol Chem 1986, 187, 1691.
24. Newman, J. K.; McCormick, C. L. Polym Prepr 1992, 33(1), 929.
25. Lee, W. F.; Tsai, C. C. Polymer 1994, 35, 2210.
26. Huggins, M. L. J Am Chem Soc 1942, 64, 2716.
27. Sakai, T. J Polym Sci Part A-2: Polym Phys 1968, 6, 1535.
28. Liaw, D. J.; Shiau, S. J.; Lee, K. R. J Appl Polym Sci 1992, 45, 61.
29. Strauss, U. P.; Leung, Y. J Am Chem Soc 1965, 1476.
30. Tan, J. S.; Marcus, P. R. J Polym Sci Polym Phys Ed 1976, 14, 239.
31. Osawa, F.; Imai, N.; Kagawa, I. J Polym Sci 1954, 13, 93.
32. Kagawa, I.; Gregor, H. P. J Polym Sci 1957, 23, 477.
33. Schulz, D. N.; Peiffer, D. G.; Agarwal, P. K.; Larabee, J.; Kaladas, J. J.; Soni, L.; Handwerker, B.; Garner, R. T. Polymer 1986, 27, 1734.
34. Lee, W. F.; Tsai, C. C. Polymer 1995, 36, 357.
35. Lee, W. F.; Tsai, C. C. J Appl Polym Sci 1994, 52, 1447.
36. Lee, W. F.; Lee, C. H. Polymer 1997, 38, 971.
37. Lee, W. F.; Hwong, G. Y. Polymer 1996, 37, 4389.
38. Chow, C. D. J Appl Polym Sci 1976, 20, 1619.
39. Lee, W. F.; Chen, Y. M. J Appl Polym Sci 2001, 80, 1619.