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Miscible blends formed from intrapolymer repulsive interactions. 2. Phase behavior of blends of bisphenol a polycarbonate and zinc-sulfonated polystyrene ionomers

Rui Xie and R.A. Weiss*
Polymer Science Program and Department of Chemical Engineering, University of Connecticut, Institute of Materials Science, 97 N. Eagleville Road, Storrs, CT 06269-3136, USA

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The phase behavior of mixtures of bisphenol A polycarbonate and the zinc salts of lightly sulfonated polystyrene ionomers (Zn-SPS) was studied as a function of the sulfonation level and the molecular weight of the ionomer. This system exhibits upper critical solution temperature (UCST) phase behavior and miscibility is attributed to intramolecular repulsion between the ionic and non-ionic groups of the ionomer. The cloud point temperature decreased with increasing sulfonation level of the ionomer over the range of 8.7–13.7 mol.%, and it increased with increasing molecular weight of Zn-SPS for a fixed sulfonation level. Fourier transform infrared spectroscopy revealed that no specific interactions involving either the carbonate carbonyl group or the metal sulfonate group occurred in the blends. The composition dependence of the glass transition of the miscible Zn-SPS/PC blends also gave no indication that specific interactions occurred in this system. © 1998 Elsevier Science Ltd. All rights reserved.

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INTRODUCTION

Numerous studies have shown that the introduction of attractive interactions between components in a polymer blend, such as hydrogen bonding[1,2], ion–dipole interactions[3], acid–base interactions[4], or transition metal complexation[5], are effective at enhancing miscibility of otherwise immiscible polymers. Compared with the large amount of research that has considered the use of specific *inter*molecular attractive interactions to enhance miscibility of otherwise immiscible polymer blends, relatively few studies have exploited *intra*polymer repulsive interactions between the different species in a random copolymer to develop miscible blends[6–10]. In the latter case, polymer mixing dilutes the very unfavorable interactions between the segments of the copolymer, and hence even though all binary segment–segment interactions in the system are repulsive, the overall interaction parameter, χ, may be negative.

Ionomers appear to be exceptional candidates for developing miscible blends with other polymers. On the one hand, the ionic group provides a rich chemistry for achieving specific interactions with polymers that contain complementary polar groups, such as amine or amide groups. On the other hand, the strong repulsive interactions that occur between the ionic and non-ionic species of the ionomer, which is in fact a random copolymer, may be an especially effective mechanism for accomplishing miscibility between polymers where no specific intermolecular interactions occur.

In a previous paper[6], we reported that blends of lightly sulfonated polystyrene ionomers (SPS) with bisphenol A polycarbonate (PC) exhibited miscibility within a specific sulfonation range that depended on the particular cation used. Differential scanning calorimetry studies also indicated that the blends exhibited upper critical solution temperature (UCST) phase behavior, which is unusual for blends of high molecular weight polymers. No interpolymer interactions were detected by Fourier transform infrared (*FT*i.r.) spectroscopy, and the miscibility was attributed to intrapolymer repulsive interactions within the SPS ionomer. In the present paper, we report phase diagrams for blends of PC and the zinc salt of SPS (Zn-SPS) measured from light scattering and optical microscopy, and a confirmation of the absence of specific interpolymer interactions using temperature-resolved *FT*i.r. **spectroscopy. The composition dependence of the glass transition temperature** (T_g) measured by differential scanning calorimetry (d.s.c.) was also consistent with that of a blend lacking strong intermolecular interactions.

EXPERIMENTAL SECTION

Materials

Bisphenol-A polycarbonate (PC), $M_n = 48\,000$ g mol^{-1}, was obtained from General Electric Co. and was used as received. SPS was prepared by sulfonating polystyrene (PS) dissolved in dichloroethane using acetyl sulfate[11]. The sulfonation reaction substitutes a sulfonic acid group at the *para*-position of the phenyl ring and sulfonation occurs randomly along the chain. Three different anionically polymerized PS with relatively narrow molecular weight distributions were used (see *Table 1*). The sulfonation level was determined by titration of the sulfonic acid derivative of the SPS product in a mixed solvent of toluene/methanol (90/

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* To whom correspondence should be addressed

Table 1 Characteristics of the zinc-sulfonated polystyrene

Sample	Sulfonation of the ionmer (mol.%)	M_w of starting PS ($g\ mol^{-1}$)	M_w/M_n
8.7Zn-SPS	8.7	4000	1.06
10.3Zn-SPS	10.3	4000	1.06
13.7Zn-SPS	13.7	4000	1.06
5.88Zn-SPS	5.88	135 000	1.13
5.89Zn-SPS	5.89	280 000	1.09

10, v/v) with methanolic sodium hydroxide. The zinc salt (Zn-SPS) was prepared by neutralizing the acid derivative in toluene/methanol (90/10) with a stoichiometric quantity of a methanol solution of zinc acetate dihydrate. The nomenclature used for the ionomers is *x.y*Zn-SPS, where *x.y* denotes the sulfonation level in mol.% of sulfonated styrene. The ionomer samples used in this study are summarized in *Table 1*.

Sample preparation

Blends were prepared by solution mixing the two polymers in a common solvent. PC was first dissolved in tetrahydrofuran (THF) to form a 1% (g/100 ml) solution, after which a small amount of methanol was added to achieve a ratio of THF and methanol of 10/1 (v/v). The PC solution was then added dropwise to a stirred 1% solution of Zn-SPS in a mixture of THF and methanol of from 10/1 to 5/1 (v/v). Blend samples for d.s.c. measurements were solution cast at 30°C. The solvent was allowed to evaporate over 3 days, and then the sample was dried in a vacuum oven at 80°C for another 3 days. Thin films of the blends for *FT*i.r. spectroscopy and optical microscopy measurements were cast from solution onto KBr or glass plates, respectively, and dried under vacuum at 80°C for 3 days.

Measurements

Cloud point temperatures, T_{cp}, were measured with a custom-built light-scattering apparatus that measured the back-scattered laser light intensity from the melt–air interface of a melt sample that was subjected to a low-amplitude oscillation[12]. The sample was heated at a rate of *ca.* 1°C min^{-1}. The back-scattered light intensity decreased as the sample was heated through the cloud point temperature (T_{cp}), and T_{cp} was defined as the intersection formed from the scattering base line below T_{cp} and the decreasing light scattering above the cloud point.

Optical micrographs of the blend morphology were obtained at various temperatures with a Nikon microscope equipped with a hot stage with temperature control of ±0.5°C. T_{cp} was also estimated by direct observation of the morphology with the microscope while heating the sample. The cloud point temperature was first estimated by heating from room temperature, where two phases were observed, at a heating rate of 20°C min^{-1} until mixing of the phases was observed. T_{cp} was then more precisely measured by heating at a rate of 1°C min^{-1} over a smaller temperature interval near the phase transition. T_{cp} was defined as the temperature where the dispersed phase began to disappear. The disappearance/appearance of a second phase was reproducible when the temperature was increased or decreased through the cloud point. The difference in T_{cp} measured from a heating or cooling experiment was generally *ca.* 3–5°C, and the values were in qualitative agreement with T_{cp} determined from light scattering.

Glass transition temperatures, T_g, were measured with a Perkin-Elmer DSC-7 using a dry nitrogen atmosphere and a heating rate of 20°C min^{-1}. The glass transition temperature was defined as the midpoint of the change in the heat capacity for the transition. Miscibility of the blends at various temperatures was assessed by annealing the blend at the desired temperature under nitrogen for 30–60 min, quenching the sample rapidly to 0°C and running a d.s.c. heating experiment. The cooling rate was assumed to be fast enough so that the subsequent heating thermogram represented the state of the blend at the annealing temperature. The blend was considered to be miscible at the annealing temperature if the subsequent heating thermogram exhibited a single T_g and immiscible if the thermogram showed two T_g values.

Fourier transform infrared (*FT*i.r.) was used to determine whether any specific interpolymer interactions involved the carbonate or the sulfonate groups. *FT*i.r. spectra were measured with a Mattson Cygnus 100 FT-IR spectrometer using a resolution of 1 cm^{-1}. A total of 320 scans were signal averaged for each spectrum. A temperature-controlled cell (± 1°C) was used for measuring spectra over the range of 25–300°C.

Micro-i.r. measurements of phase-separated blends were made with a Spectra Tech IR-Plan™ Microscope accessory interfaced with a Nicolet 60SX FTIR. Spectra were obtained on ~10 μm diameter area using 128 scans with a resolution of 4 cm^{-1}. The samples for micro-i.r. were prepared by solution-casting polymer films onto 2-mm thick BaF_2 windows.

RESULTS AND DISCUSSION

Figure 1a shows an optical micrograph of a 10.3Zn-SPS/PC blend containing 80 wt.% ionomer and 20 wt.% PC after casting a film from solution, and *Figure 1b* is the same film after annealing at 170°C under vacuum for 24 h. The as-cast film was phase-separated, as were cast films for all the other blend compositions. The absence of birefringence when the films were observed between cross-polarizers confirmed that the dispersed phase in *Figure 1a* was not due to crystallization of the PC, which may occur when PC is isolated from solution. Although the contrast between the two phases was sufficient for resolution with an optical microscope, the films appeared relatively clear. The difference in the refractive indices (Δn) for PS and PC is less than 0.002[13], which minimizes the light scattering from a two-phase material. However, sulfonation of the PS appeared to increase Δn sufficiently so that the cloud point curves of the ionomer/PC blends were measurable by light scattering. *Figure 2* shows the micro-i.r. measurements taken of one of the phase-separated domains (area A) and of the surrounding continuous phase (area B). The spectra indicate that the domains are pure polycarbonate and the continuous phase is rich in the ionomer, but contains some polycarbonate. The composition of the continuous phase was estimated to be *ca.* 94% (wt.) Zn-SPS and 6% PC by comparing the ratio of the bands at 1773 (PC) and 1450 cm^{-1} (PS) with a calibration curve constructed from miscible blends of known compositions.

The solvent-cast films were annealed for 24 h under vacuum at 170°C, which is above the T_g of both polymers, in order to confirm that the phase-separation in *Figure 1a* was not a consequence of the solvent-casting procedure. That thermal treatment had no effect on the morphology of the blend, as shown in *Figure 1b* for the (80/20) 10.3Zn-SPS/PC blend. That result indicates that the two-phase morphology observed at room temperature for the as-cast films was the equilibrium morphology, and was not a consequence of

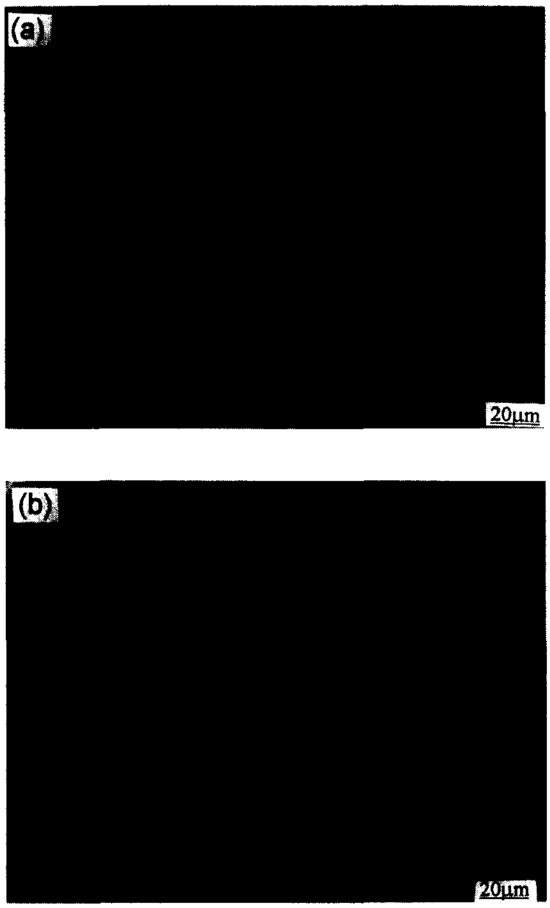

Figure 1 Optical micrographs for a blend of 10.3Zn-SPS/PC containing 20 wt.% PC: (a) film cast from solution; (b) film annealed at 170°C after casting from solution

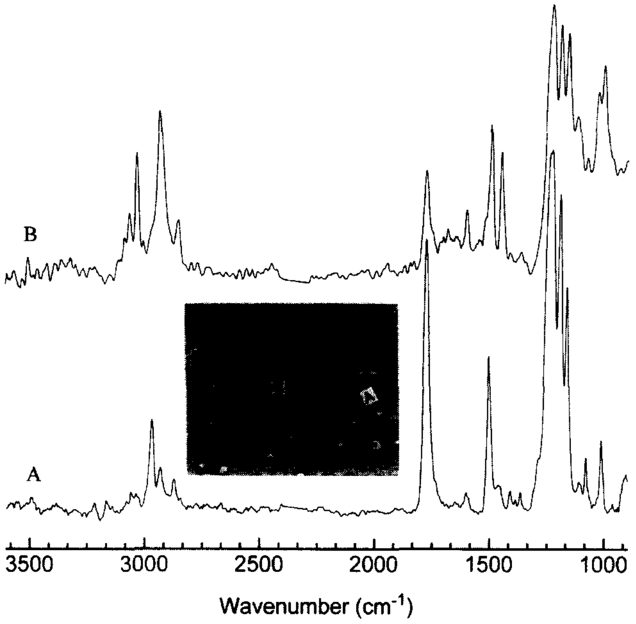

Figure 2 Micro-i.r. spectra for (80/20) 10.3Zn-SPS/PC blend. The bottom spectrum is of the phase-separated domain, A, and the top spectrum is from area B of the optical micrograph

solvent casting. That conclusion is supported by the fact that the Zn-SPS/PC blends exhibited upper critical solution temperature (UCST) phase behavior in contrast to blends in which strong intermolecular interactions occur, such as in SPS/PA6 blends that exhibit lower critical solution temperature (LCST) phase behavior[14]. Therefore, the

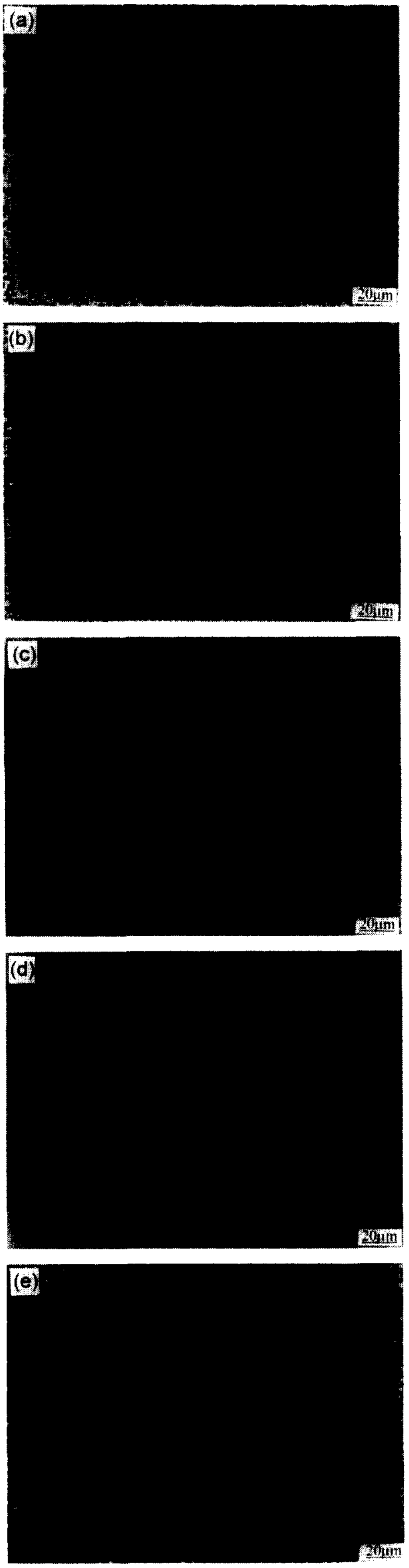

Figure 3 Optical micrographs for a solution-cast film of a blend of a (50/50) 13.7Zn-SPS/PC blend at: (a) 200, (b) 225, (c) 250, and (d) 250°C after annealing for 1 h; and (e) after cooling from 250 to 200°C and annealing for 1 h

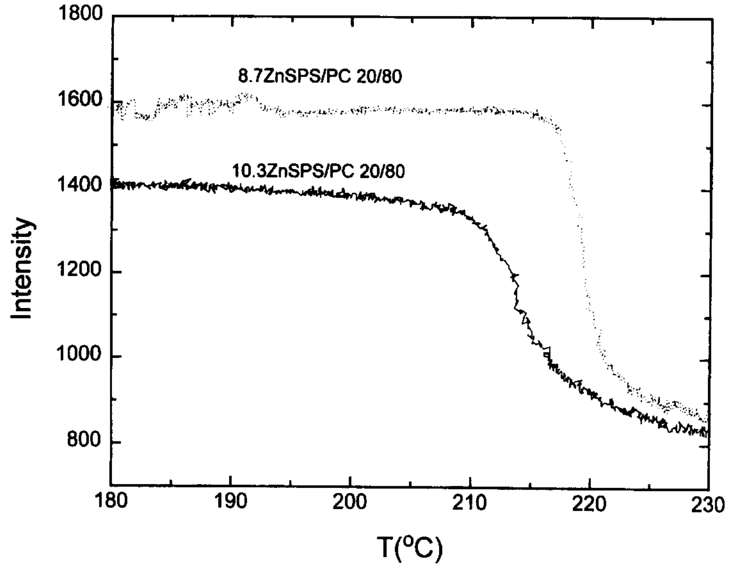

Figure 4 Back-scattered light scattering intensity *versus* temperature for two Zn-SPS/PC blends. $M_w = 4000\ \mathrm{g\ mol^{-1}}$ for the ionomer

phase separation shown in *Figure 1* is also not due to annealing the blend at elevated temperature.

Phase mixing of the blends was observed when the samples were heated above the UCST. Optical micrographs for a (50/50) 13.7Zn-SPS/PC blend following different thermal histories are shown in *Figure 3*. The two-phase morphology observed at room temperature persisted to 200°C (*Figure 3a*), but mixing of the phases began when the temperature was raised to 225°C (*Figure 3b*). The temperature was raised to 250°C to accelerate the mixing rate (*Figure 3c*), and after 1 h at 250°C, only a single, homogeneous phase was observed (*Figure 3d*). The UCST phase behavior was reversible as shown in *Figure 3e*, which is a micrograph of the same sample as in *Figure 3d* after cooling from 250 to 200°C and annealing for 1 h. Similar results were observed for blends with different compositions and/or when the sulfonation level of the ionomer was varied.

The phase diagrams of the various Zn-SPS/PC blends were determined by light scattering. Typical light scattering data obtained during heating for two different blends are shown in *Figure 4*, and the cloud-point curves measured for blends using ionomers with different molecular weights and sulfonation levels are shown in *Figure 5*. In *Figure 5a*, the molecular weight of the ionomer used in the three blends was held constant at $M_w = 4000\ \mathrm{g\ mol^{-1}}$, but the sulfonation level was varied from 8.7 to 13.7 mol.%. Although the SPS molecular weight used for the blends in *Figure 5a* was relatively low, blends of PC and the starting PS were completely immiscible over a comparable temperature range. All the blends exhibited UCST phase behavior, and T_{cp} decreased with increasing sulfonation of the ionomer. That result is in stark contrast with the effect of strong intermolecular interactions on the phase behavior of blends, such as SPS/polyamide blends, which exhibit LCST-type phase behavior and a cloud point that increases with increasing sulfonation level[14–16]. The decrease of T_{cp} with increasing sulfonation of the ionomer for the Zn-SPS/PC blends is a consequence of the increasing miscibility of the polymers as the concentration of intra-ionomer repulsive interactions increases. The critical point for the blends (see *Figure 5a*) was skewed towards the higher molecular weight component, i.e. PC, which is what is usually expected in partially miscible blends, though the critical composition, *ca.* 60 wt.% PC, was relatively insensitive to the sulfonation level.

Blends of Zn-SPS/PC based on higher molecular weight ionomers also exhibited UCST-phase behavior. *Figure 5b* shows the effect of the SPS molecular weight on the phase behavior of Zn-SPS/PC blends for a constant sulfonation level of *ca.* 5.9 mol.%. M_w of the ionomer was varied from 135 000 to 280 000 g mol^{-1}. For a partially miscible binary blend, increasing the molecular weight of either component should reduce the miscibility, and *Figure 5b* shows that UCSTs of the Zn-SPS/PC blends increased when the molecular weight of the SPS was doubled. A comparison of *Figure 5a* and *Figure 5b* indicates that the critical composition shifted from PC rich to ionomer rich, i.e. from 60 to 40% PC, as the M_w of the ionomer increased from 4000 to 135 000 g mol^{-1}, which again is an expected result as the higher molecular weight component switches from one polymer to the other.

The T_g behavior of the blends following different thermal histories were consistent with the phase diagrams shown in *Figure 5*. D.s.c. heating thermograms (not shown) following temperature quenches to room temperature for samples annealed below the T_{cp} shown in *Figure 5* showed two T_g values, which indicates a phase-separated system at the annealing temperature. The lower T_g corresponded to the glass transition of a Zn-SPS-rich phase, and the higher T_g corresponded to the glass transition of a PC-rich phase. For the as-cast blends, the two T_g values were close to those of the pure component polymers, which indicates that the two phases were nearly pure ionomer and PC. That result might be expected if the phase diagrams in *Figure 5b* were extrapolated to room temperature. However, for samples annealed in the two-phase region, but nearer the UCST, the two T_g values were shifted towards each other, which indicates some mixing of the polymers in each phase. Again, that is consistent with the phase diagrams in *Figure 5*. For example, the cloud point curve in *Figure 5b* for the 5.9Zn-SPS/PC blend using the $M_w = 280\,000\ \mathrm{g\ mol^{-1}}$ ionomer indicates that at 230°C the two phases contain 20 and 80% PC, respectively. When the blends were annealed above the T_{cp} read from *Figure 5*, the d.s.c. thermograms following a temperature quench to room temperature showed only a single T_g intermediate between the T_g

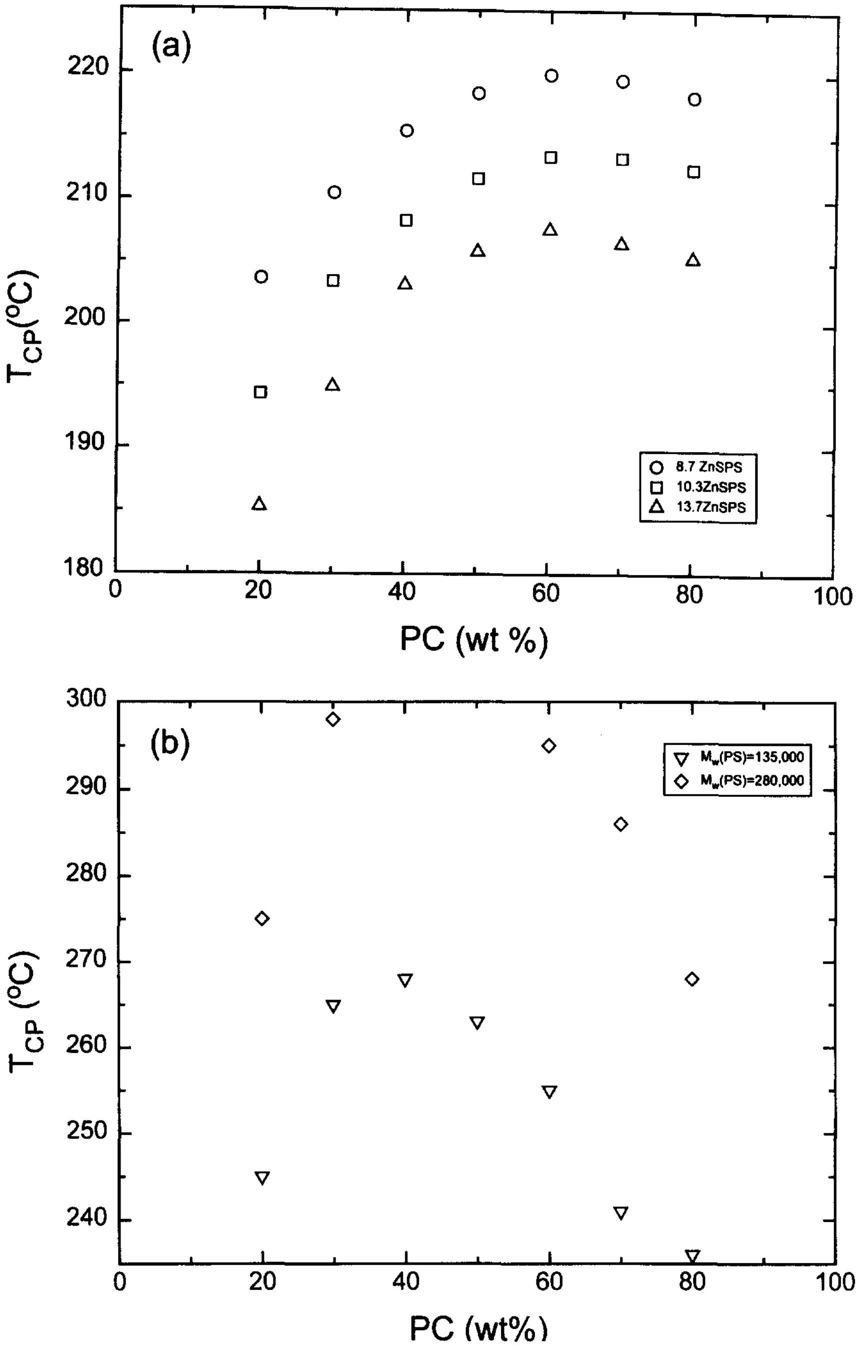

Figure 5 Cloud point curves determined by light scattering for blends of Zn-SPS and PC as a function of: (a) the sulfonation level (M_w = 4000 g mol^{-1} for the ionomer) and (b) the ionomer molecular weight (sulfonation level of the ionomer = 5.9 mol.%)

values of the pure component polymers, which is characteristic of miscible blends.

Possible degradation of the polymers was one concern that we had with the high temperatures used to develop the phase diagrams and the long annealing times, e.g., 30 min, that were sometimes used to allow the morphologies to equilibrate. Lowering of the molecular weight of the polymers at the high temperatures could perhaps be responsible for the enhanced miscibility of the polymers, and give the false impression of a UCST. However, measurements of the inherent viscosity of a 50/50 blend annealed for 30 min at 275°C indicated little if any degradation of the polymers occurred. The inherent viscosity as measured with a Ubbelohde capillary viscometer using a total polymer concentration of 0.445 g dl^{-1} was 0.215 and 0.212 dl g^{-1}, respectively, before and after that annealing procedure. As a result, it is unlikely that degradation of the polymers is responsible for the phase behavior of the ionomer/PC blends reported in this paper.

The T_g values of the 5.9Zn-SPS/PC blends obtained after annealing at a temperature within the single-phase region, i.e. above the T_{cp} read from *Figure 5b*, are plotted against the blend composition in *Figure 6*. The T_g *versus* composition curve for these blends, as well as for the other miscible blends investigated in this study, showed negative deviation from a simple, weighted average of the two component polymer T_g values. For very weak or no specific intermolecular interactions, i.e. $\chi \sim 0$, the Gordon–Taylor[17] equation adequately predicts the T_g for miscible polymer blends. The predictions of the Gordon–Taylor equation for the Zn-SPS/PC blends are shown by the solid lines in *Figure 6*. The agreement of the Gordon–Taylor

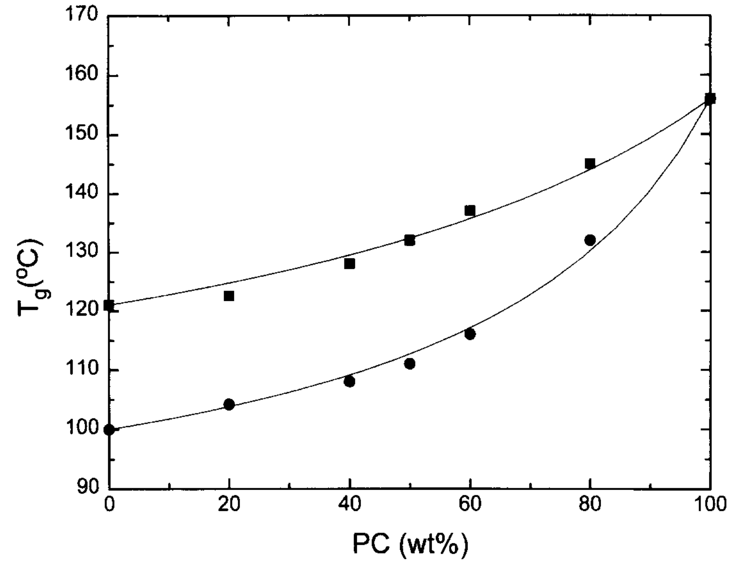

Figure 6 T_g *versus* composition for 10.3Zn-SPS/PC (●) and 5.9Zn-SPS/PC (■) blends. The solid lines are the fits of the Gordon–Taylor equation[17]

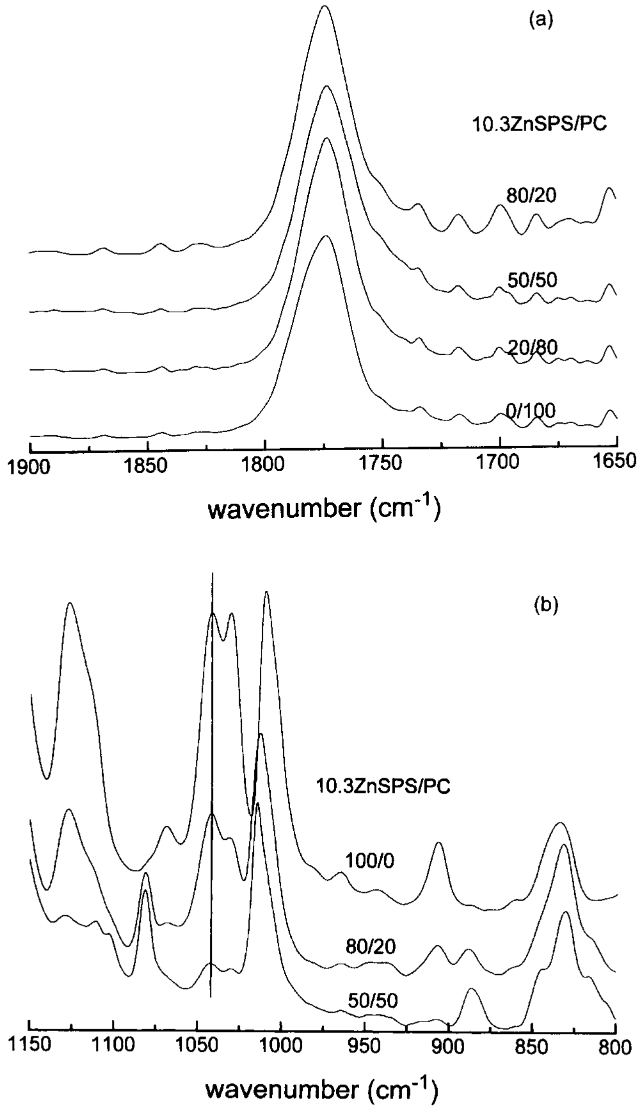

Figure 7 Room-temperature *FT*i.r. spectra of (a) the carbonate carbonyl stretching region and (b) the sulfonate stretching region for the 10.3Zn-SPS/PC blends. Blend compositions are indicated in the figure

equation with the data indicates that any specific interaction that may occur between Zn-SPS and PC is weak—and certainly much weaker than the intermolecular interactions that occur in Zn-SPS/polyamide blends[15,16], which exhibit positive deviation of T_g from a linear weighted average of the component T_g values.

The absence of specific interactions between the carbonate and metal sulfonate groups was confirmed by *FT*i.r. analyses. *Figure 7a* shows the *FT*-i.r. spectra in the carbonyl stretching region for solution-cast samples of the neat PC and several compositions of the 10.3Zn-SPS/PC blends. For clarity, the different spectra are shifted vertically in *Figure 7*. The position and shape of the absorption at 1773 cm^{-1} due to the stretching vibration of the PC carbonyl groups did not change upon addition of Zn-SPS. Similar results were obtained for the other blends studied. *Figure 7b* shows the sulfonate stretching region, 800–1150 cm^{-1}, for the neat 10.3Zn-SPS and several 10.3Zn-SPS/PC blends. The absorption band at 1044 cm^{-1}, due to the symmetric S–O stretching vibration of the SO_3^- anion with a Zn^{2+} cation, is very sensitive to the local environment of the sulfonate anion. Any weakening of the ion-pair interaction, such as would occur if the cation and/or the anion were solvated or complexed by another chemical species, e.g., the carbonate carbonyl group, should perturb the S–O vibration frequency. As demonstrated by the invariance of the 1044-cm^{-1} band for the neat ionomer and the blends in *Figure 7b*, no specific interaction occurs between the sulfonate group and PC in the solution cast blends.

As discussed above, the two phases in the solution-cast blends were nearly pure component polymers, so the absence of an intermolecular interaction between the sulfonate and carbonate groups in *Figure 7* does not preclude that specific interactions may occur above T_{cp}, i.e. in the miscible blends. *Figure 8* shows the *FT*i.r. spectra of the carbonyl and sulfonate stretching regions for a (50/50) 10.3Zn-SPS/PC blend measured at 200, 235 and 250°C, which correspond to temperatures below, near and above T_{cp}, respectively. In each case, the spectrum was recorded after the film was annealed at the indicated temperatures for at least 30 min in order to equilibrate the morphology. Although there is some broadening of the carbonyl peak at

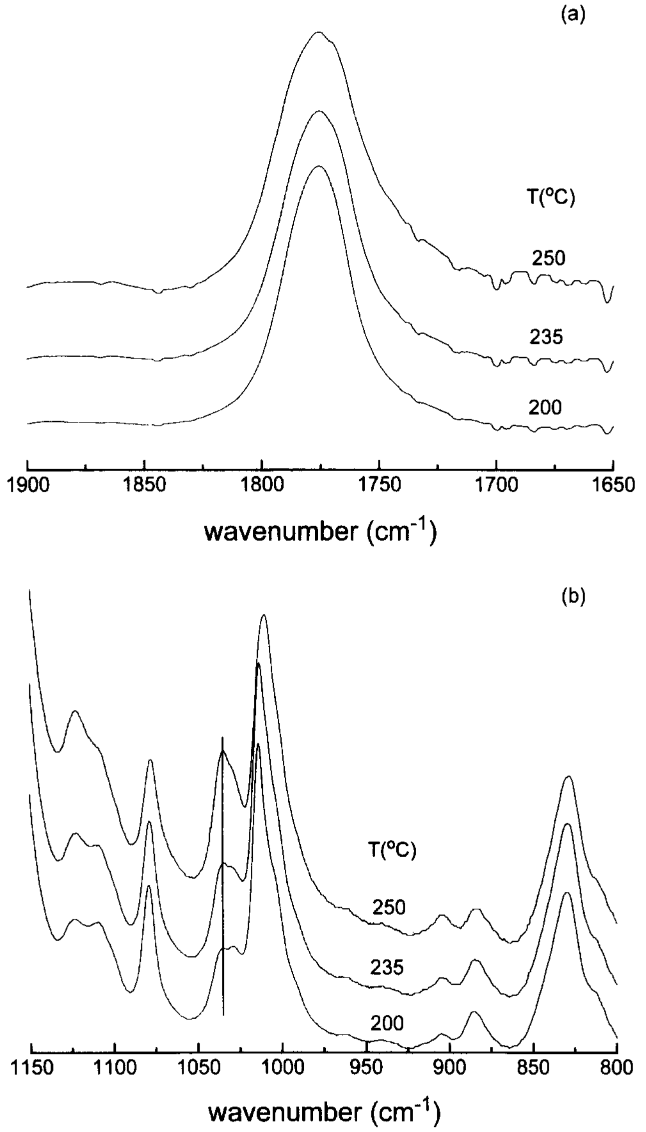

Figure 8 Temperature-resolved *FT*i.r. spectra for a (50/50) 10.3Zn-SPS/PC blend: (a) carbonyl stretching region and (b) sulfonate stretching region

the highest temperature, the maximum of the carbonate carbonyl absorption remained constant over the temperature range studied. Similarly, the absorption peak for the symmetric S–O stretching vibration of the sulfonate anion did not shift with increasing temperature, though there appears to be some changes to a lower frequency neighboring vibration. The constancy of the frequency of the carbonyl and S–O stretching vibration from room temperature to 250°C indicates that no significant change in the environment of both groups occurred over that temperature interval, which supports the conclusion that no specific interaction occurs between Zn-SPS and PC.

The absence of specific intermolecular interactions and the observation of a UCST phase diagram for the Zn-SPS/PC blend strongly suggests that intrapolymer repulsive interactions, i.e. the copolymer effect[7–9], are responsible for miscibility of these polymers. Although miscibility due to the copolymer effect is not unique for SPS and PC, it is significant in that the repulsive intramolecular interaction in an ionomer between a salt and an uncharged species is expected to be much stronger than the dispersive interactions usually found in random copolymers of non-ionic monomers. Although not specifically demonstrated in the present paper, the magnitude of the repulsive interaction effect may be controlled to some extent by varying the ionomer counterion, and this may be used to adjust the phase behavior[6].

CONCLUSIONS

Blends of polycarbonate and the zinc salt of lightly sulfonated polystyrene ionomers are partially miscible and exhibit UCST phase behavior. For fixed molecular weights, the cloud point temperature decreases with increasing sulfonation level of the ionomer, and for a fixed sulfonation level, the cloud point increases with increasing molecular weight of the ionomer. The T_g-composition behavior of miscible Zn-SPS/PC blends exhibits negative deviation from a linear weighted average of the component polymer T_g values, which is consistent with a miscible blend with weak or no specific interpolymer interactions. *FT*i.r. spectroscopy failed to identify any perturbation of the carbonate carbonyl or the metal sulfonate anion vibrations as a result of blending the ionomer and PC, which supports the conclusion of no specific intermolecular interaction in this system. These results are consistent with previously published results for blends of PC with other salts of SPS ionomers[6], and they strongly suggest that the miscibility between these two polymers arises from intrapolymer repulsive interactions between the ionic and non-ionic species of the ionomer.

These results also suggest that miscibility of ionomers with other polymers, even when no specific intermolecular interactions occur, may be a common phenomenon. Although further research on additional systems is necessary to confirm that hypothesis, they also suggest the versatility of ionomers as compatibilizing agents for a wide variety of polymer blends, including those in which interpolymer interactions are possible and those in which they do not occur[18].

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