

# Compatibility Studies of Sulfonated Poly(ether ether ketone)–Poly(ether imide)–Polycarbonate Ternary Blends

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**ABSTRACT:** Binary blends of the sulfonated poly(ether ether ketone) (SPEEK)–poly(ether imide) (PEI) and SPEEK–polycarbonate (PC), and ternary blends of the SPEEK–PEI–PC, were investigated by differential scanning calorimetry. SPEEK was obtained by sulfonation of poly(ether ether ketone) using 95% sulfuric acid. From the thermal analysis of the SPEEK–PEI blends, single glass transition temperature ( $T_g$ ) was observed at all the blend composition. For the SPEEK–PC blends, double  $T_g$ s were observed. From the results of thermal analysis, it is suggested that the SPEEK–PEI blends are miscible and the SPEEK–PC blends are immiscible. Polymer–polymer interaction parameter ( $\chi_{12}$ ) of the SPEEK–PEI blends was calculated from the modified Lu and Weiss equation, and found to range from  $-0.011$  to  $-0.825$  with the blend composition. For the SPEEK–PC blends, the  $\chi_{12}$  values were calculated from the modified Flory–Huggins equation, and found to range from  $0.191$  to  $0.272$  with the blend composition. For the SPEEK–PEI–PC ternary blends, phase separation regions that showed two  $T_g$ s were found to be consistent with the spinodal curves calculated from the  $\chi_{12}$  values of the three binary blends. © 2000 John Wiley & Sons, Inc. *J Appl Polym Sci* 78: 2488–2494, 2000

**Key words:** sulfonated poly(ether ether ketone); poly(ether imide); polycarbonate; blends; compatibility

## INTRODUCTION

Blends of miscible polymers as well as partially miscible polymers typically have certain advantages. In partially miscible blends, phase separation occurs, but at the same time a certain number of molecules of one phase penetrate the other phase and vice versa.<sup>1–3</sup> The interface between the two phases becomes diffuse and good mechan-

ical properties may result by the introduction of secondary intermolecular interactions such as hydrogen bonding, ionic interactions, and charge transfer complex.<sup>4–6</sup> In order to modify the chemical structure of polymer, there are several methods such as sulfonation, chlorination, and nitration of polymers, etc. Among these methods, sulfonation of polymers, which is a versatile method to obtain processable ether ketones,<sup>7</sup> has been investigated in recent years.<sup>8–10</sup>

Karcha and Porter<sup>11,12</sup> studied the miscibility of modified poly(aryl ether ketone) with aromatic polyimides. From a spectroscopic investigation of the blends, they reported that the blends are miscible at all the blend compositions.<sup>11</sup> The misci-

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**Table I** Characteristics of Polymer Samples Used in This Study

	$\overline{M}_w^a$	$\overline{M}_n^a$	$T_g$ (°C) <sup>b</sup>	$\Delta C_p$ (J/g°C) <sup>b</sup>	$T_m$ (°C) <sup>b</sup>	$\rho$ (g/cm <sup>3</sup> ) <sup>c</sup>
PEEK <sup>d</sup>	39400	14000	146.0	0.308	340.0	1.26
PEI <sup>e</sup>	30000	12000	218.9	0.241	—	1.27
PC <sup>f</sup>	24100	10000	150.9	0.226	—	1.20

<sup>a</sup> Measured in our laboratory by gel permeation chromatography.

<sup>b</sup> Measured in our laboratory by DSC.

<sup>c</sup> Measured in our laboratory by specific gravity chain balance.

<sup>d</sup> Supplied by ICI.

<sup>e</sup> Supplied by General Electric Co.

<sup>f</sup> Supplied by Sam Yang Kasei Co., Ltd.

bility between two polymers is mainly responsible for the formation of electron donor–acceptor complexes between the sulfonated/sulfamidated phenylene rings of the poly(ether ether ketone) (PEEK) and the N-phenylene units of the polyimides.<sup>12</sup> For the study of phase behavior of sulfonated PEEK (SPEEK)–poly(ether imide) (PEI)–poly(amide imide) (PAI) ternary blends, they reported that SPEEK in the SPEEK–PEI–PAI ternary blends can act as a compatibilizing agent between the PEI and PAI.<sup>12</sup> Recently, we have shown that the blends of PEEK and PEI are miscible in the amorphous state and partially miscible in the semicrystalline state.<sup>13,14</sup>

The method of determining the polymer–polymer interaction parameter between component polymers in a miscible blend has been studied widely.<sup>15–23</sup> Several techniques can be used to determine the thermodynamic polymer–polymer interaction parameter ( $\chi_{12}$ ), such as melting point depression,<sup>15,16</sup> vapor sorption,<sup>17</sup> inverse-phase gas chromatography,<sup>18,19</sup> some light-scattering methods,<sup>20,21</sup> and glass transition temperature method.<sup>14,22,23</sup> Only a few methods have been developed which can be applied to incompatible polymer blend systems.<sup>3,24–27</sup>

In this paper, we examine the thermal properties and phase behavior of binary blends of SPEEK–PEI and SPEEK–PC, and SPEEK–PEI–PC ternary blends by differential scanning calorimetry. Polymer–polymer interaction parameters of the SPEEK–PEI and SPEEK–PC blends were calculated using the thermal analysis results. From the  $\chi_{12}$  values obtained from the three binary blends, the phase behavior of the SPEEK–PEI–PC ternary blends was simulated using the modified Flory–Huggins equation, which can be applied to the ternary polymer blends.<sup>28</sup> These spinodals are compared with the  $T_g$  results obtained from the DSC measurements of the SPEEK–PEI–PC ternary blends.

## EXPERIMENTAL

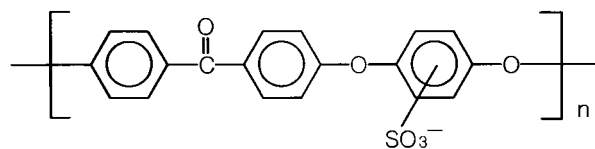
### Polymers

The polymers used in this study were obtained from commercial sources. The characteristics and sources of the PEEK, PEI, and PC are shown in Table I. The PEEK (VICTREX 450G) used in this study was provided by ICI. The PEI (Ultem 1000) was supplied by General Electric Co. The PC (TRIUREX 3025A) was supplied by Sam Yang Kasei Co., Ltd.

The SPEEK was prepared by the method previously developed by Jin et al.<sup>29</sup> For sulfonation of PEEK, the PEEK granules were dissolved in H<sub>2</sub>SO<sub>4</sub> (assay 95.0%, Junsei Chemical Co.) [5.0% (w/v) solution] and stirred at room temperature. The stirring speed was set at 130 rpm. Sulfonation time was varied from 45 to 175 h. The resulting sulfonated PEEK was precipitated by dropwise addition into six volumes of deionized H<sub>2</sub>O. The SPEEK was obtained by filtration and rigorously washed with distilled water to remove excess acid, and then dried under vacuum at 100°C. Sulfonation level of the SPEEK was determined by elemental analysis.

It is known that sulfonation does not occur on the phenylene rings adjacent to the carbonyl group but occurs on the phenylene ring between the two ether linkages.<sup>29,30</sup> The chemical structure of SPEEK is shown in Figure 1.

When the degree of sulfonation ( $X_s$ ) is below 0.5, the SPEEK was not completely soluble in



**Figure 1** Chemical structure of sulfonated poly(ether ether ketone).

N,N-dimethylacetamide (DMAc) at room temperature. Therefore, the SPEEK that was sulfonated for 65 h ( $X_s = 0.72$ ) was used in the preparation of SPEEK-PEI and SPEEK-PC blends, and SPEEK-PEI-PC ternary blends.

### Blend Preparations

The blends of SPEEK-PC, SPEEK-PEI, and SPEEK-PEI-PC were prepared by solution casting. The polymer samples were dried under vacuum at 120°C for 24 h before use. For the blends of SPEEK-PC, SPEEK-PEI, and SPEEK-PEI-PC, a total 0.6 g was dissolved in 20 mL of DMAc (Junsei Chemical Co.) [3.0% (w/v) solution]. For the PEI-PC blends, methylene chloride was used as a solvent.<sup>31</sup> These solutions were stirred at room temperature until homogeneous and cast on glass plates. To remove residual solvents, all blend films were dried under vacuum for 3 days at 160°C.

### Differential Scanning Calorimetry Measurements

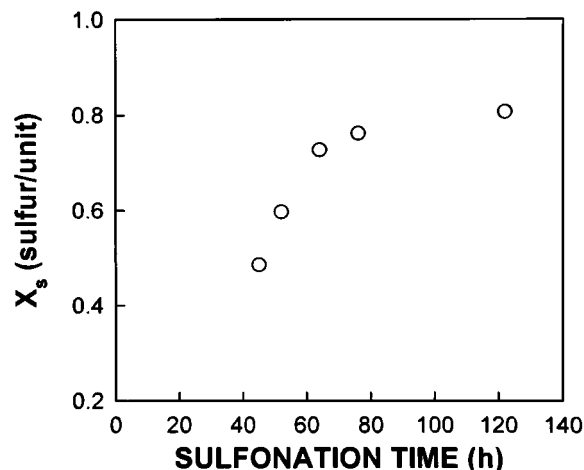
The thermal properties of all samples were measured calorimetrically using a Perkin-Elmer differential scanning calorimetry (DSC), Model DSC-7. Temperature calibration was performed using indium ( $T_m = 156.60^\circ\text{C}$ ,  $\Delta H_f = 28.5 \text{ J/g}$ ). Blend samples of 5–15 mg were heated in a nitrogen atmosphere from 50 to 260°C at a heating rate of 20 K/min and then naturally cooled to 50°C for the second scan.

## RESULTS AND DISCUSSION

### Sulfonation of PEEK

SPEEK was prepared by the method developed by Jin et al.<sup>29</sup> The effect of sulfonation time on the degree of sulfonation ( $X_s$ ) is shown in Figure 2. The degree of sulfonation is defined as the average number of sulfone group per polymer repeating unit, and determined from the sulfur/carbon ratio of each sample by elemental analysis. In Figure 2, we can see that the degree of sulfonation increases with the increase of sulfonation time. The  $X_s$  of the SPEEK that was sulfonated for 45 and 122 h was found to be 0.48 and 0.80, respectively.

The effect of sulfonation time on the glass transition temperature ( $T_g$ ) of the SPEEK is shown in Figure 3. The  $T_g$  of the SPEEK increases with the increase of sulfonation time. The  $T_g$  of SPEEK



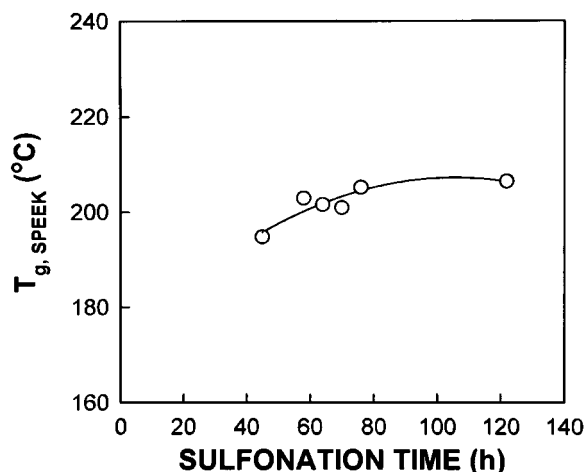
**Figure 2** Effect of sulfonation time on the degree of sulfonation ( $X_s$ ).

was determined to be 195°C (sulfonation time = 45 h) and 207°C (sulfonation time = 122 h). The increase in  $T_g$  with sulfonation time is attributed to the polar/ionic interactions involving the sulfonic acid groups and hindered rotation along the chain caused by the bulky  $-\text{SO}_3\text{H}$  groups.<sup>29,32</sup>

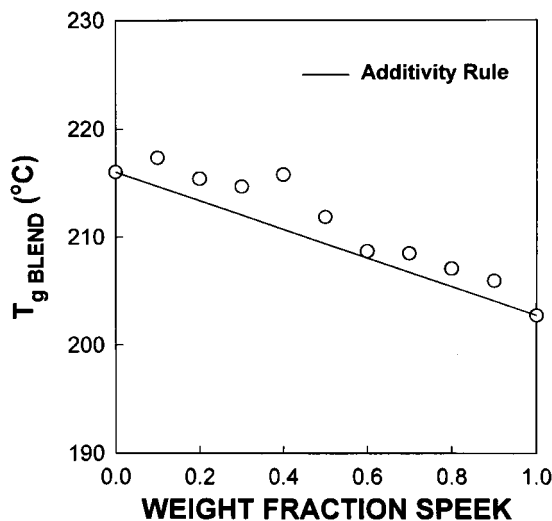
In the study of thermal analysis of SPEEK by DSC, the melting endotherm was not observed. This result indicates that the crystallization of PEEK is suppressed by the sulfonation of PEEK.

### Polymer-Polymer Interaction Parameter of SPEEK-PEI Blends

Thermal behavior of the SPEEK-PEI blends was studied using DSC. The effect of blend composi-



**Figure 3** Effect of sulfonation time on the  $T_g$  of SPEEK.



**Figure 4** Effect of blend composition on the  $T_g$  of the SPEEK-PEI blends. The curve represents the linear additivity rule.

tion on the  $T_g$  of the SPEEK-PEI blends is shown in Figure 4. A single  $T_g$ , indicating the miscibility between SPEEK and PEI, was observed for all the blend composition. In Figure 4, we can see that the experimental  $T_g$  values is higher than the values expected from the linear mixing rule. This positive deviation in  $T_g$  from the linear additivity is often cited as an indication of strong intermolecular interactions in the blends.<sup>22</sup> Karcha and Porter<sup>11</sup> studied the miscibility of SPEEK with aromatic polyimides. From spectroscopic investigations, they reported that formation of electron donor-acceptor complex between the sulfonated phenylene rings of the SPEEK and the N-phenylene units are responsible for the miscibility.

The values of the specific heat increment ( $\Delta C_p$ ) at  $T_g$  of the SPEEK-PEI blends are shown in Figure 5. From this figure, we can see that the  $\Delta C_p$  of SPEEK-PEI blends decreases with an increase of the SPEEK weight fraction and is smaller than the  $\Delta C_p$ , which is expected on the basis of the simple additivity rule except for the 0.8 and 0.9 weight fraction SPEEK.

From the difference in the  $\Delta C_p$  values between the experimentally determined  $\Delta C_p$  of the blends and the  $\Delta C_p$  from the simple additivity rule, it is found that the excess heat capacities of mixing have negative values. The negative excess heat capacities of mixing were observed in the miscible blends by other researchers.<sup>33,34</sup> Such as in poly( $\epsilon$ -caprolactone)-polychlorostyrene by Allard and Prud'homme.<sup>33</sup> Wang and co-workers<sup>34</sup> ob-

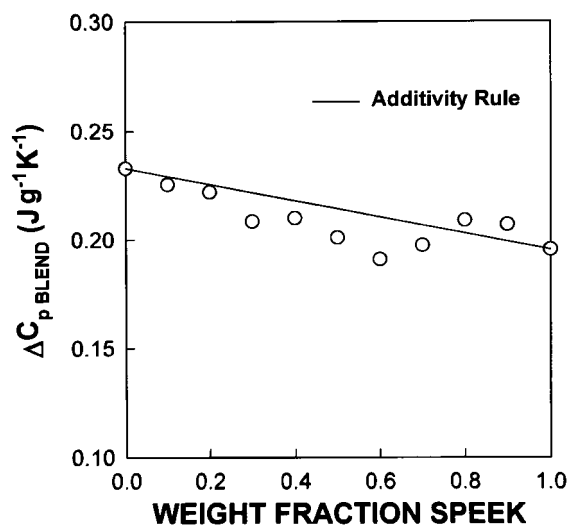
served the negative excess heat capacities of mixing for the several miscible blends having hydrogen bonding between the two polymers. From the results of  $\Delta C_p$  of the SPEEK-PEI blends, it is suggested that there is a favorable interaction between the SPEEK and PEI in the blends from the results of the negative excess heat capacities and the single glass transition temperature of the SPEEK-PEI blends, which is similar to the results of other researcher.<sup>11</sup>

Lu and Weiss<sup>22,23</sup> derived the relationship between the glass transition temperature and the interaction parameter of miscible binary polymer blends. Recently, Chun et al.<sup>14</sup> modified the Lu and Weiss equation by using the glass transition temperature as follows:

$$T_{gm} = \frac{w_1 T_{g1} + k w_2 T_{g2}}{w_1 + k w_2} + \frac{A w_1 w_2}{(w_1 + k w_2)(w_1 + b w_2)(w_1 + c w_2)^2} + \frac{k w_2 \Delta T_2 - w_1 \Delta T_1}{8(w_1 + k w_2)} \quad (1)$$

$$A = \frac{\chi_{12} R (T_{g1} - T_{g2}) c}{M_1 \Delta C_{p1}} \quad (2)$$

$$k = \frac{\Delta C_{p2} - w_1 \delta C_p^1}{\Delta C_{p1} - w_2 \delta C_p^g} \quad (3)$$



**Figure 5** Specific heat increment ( $\Delta C_p$ ) at the  $T_g$  of the SPEEK-PEI blends. The curve represents the linear additivity rule.

where  $T_{gm}$  is the observed  $T_g$  of the blend,  $w_1$  is the weight fraction of polymer 1 having  $T_{g1}$ , and  $w_2$  is the weight fraction of polymer 2 having  $T_{g2}$ . The  $b = M_2/M_1$ , where  $M_1$  and  $M_2$  are the molecular weight of the repeating unit in polymers 1 and 2, respectively.  $c = \rho_1/\rho_2$ , where  $\rho_1$  and  $\rho_2$  are the density of the components 1 and 2, respectively. The  $\chi_{12}$  is the Flory–Huggins polymer–polymer interaction parameter.  $\Delta C_p = C_p^1(T_g) - C_p^g(T_g)$ ; the specific heat increment at  $T_g$  and  $\delta C_p$  is the specific heat change due to mixing. Because  $\delta C_p$  is usually small compared with  $\Delta C_p$ , the right side of eq. (3) can be replaced by  $\Delta C_{p2}/\Delta C_{p1}$ .

The  $\chi_{12}$  values calculated by eq. (1) are not segmental  $\chi_{12}$  values. The segmental  $\chi_{12}$  values can be defined by using the following eq. (4)<sup>14</sup>:

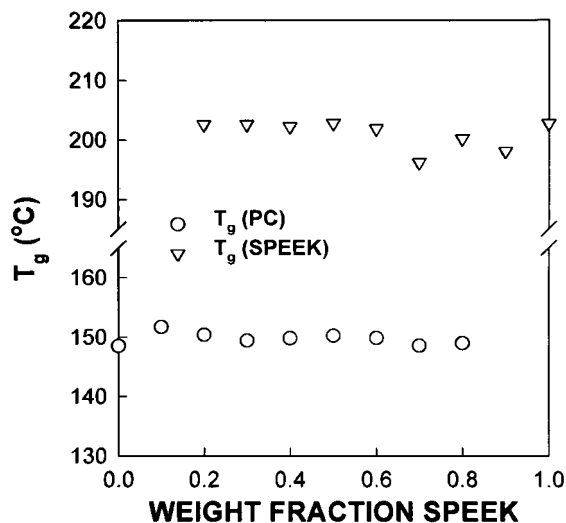
$$\Delta H_m(T) = \chi_{12}RT\phi_1\phi_2(m_1n_1 + m_2n_2) \quad (4)$$

where  $\phi_i$  is the volume fraction of component  $i$ . The  $n_1$  and  $n_2$  are the number of moles of the SPEEK and PEI in the blend, respectively. The  $m_1$  and  $m_2$  are the number of lattice site of the SPEEK and PEI molecules, respectively. The  $m_1$  and  $m_2$  can be obtained by the following relation<sup>3,24–27</sup>:  $m_1 = V_1/V_0$  and  $m_2 = V_2/V_0$ , where  $V_1$ ,  $V_2$ , and  $V_0$  are the molar volume of SPEEK and PEI, and the repeating unit of PEI, respectively. The segmental  $\chi_{12}$  values of the SPEEK–PEI blends can be obtained by dividing the  $\chi_{12}$  values calculated from eq. (1) by the average values of  $m_1$  and  $m_2$ .<sup>14</sup> The values of  $m_1 = 22.3$  and  $m_2 = 20.3$  were used for the SPEEK and PEI, respectively. A repeating unit of the PEI has been chosen as a site volume.

In Table II, the segmental  $\chi_{12}$  values of the SPEEK–PEI blends that are calculated from eq. (1) are presented. From Table II, the  $\chi_{12}$  values of the SPEEK–PEI blends are found to range from  $-0.011$  to  $-0.825$  with the blend composition.

#### Polymer–Polymer Interaction Parameter of SPEEK–PC and PEI–PC Blends

The effect of blend composition on the  $T_g$  of SPEEK–PC blends is shown in Figure 6. Two  $T_g$ s, indicating phase separation between SPEEK and PC are observed for the various blend compositions. In Figure 6, a maximum decrease of  $T_g$  of SPEEK (about 6 K) was observed in the SPEEK–PC blends. The  $T_g$  of PC in the SPEEK–PC blends is shown to almost unchanged compared to that of the pure PC.



**Figure 6** Effect of blend composition on the  $T_g$  of the SPEEK and PC in the SPEEK–PC blends: (▽)  $T_g$  of SPEEK and (○)  $T_g$  of PC.

From the  $T_g$ (SPEEK) and  $T_g$ (PC) in the SPEEK–PC blends, we can estimate the apparent weight fraction of SPEEK and PC dissolved in the PC-rich phase and the SPEEK-rich phase, respectively. The apparent weight fractions were determined by the Fox equation,<sup>35</sup> which is often used to describe the dependence of  $T_g$  on composition in miscible blend system.<sup>3,24–27</sup> From the apparent weight fractions of SPEEK and PC in the SPEEK-rich phase and in the PC-rich phase, we can calculate the apparent volume fractions of SPEEK and PC in the SPEEK-rich phase and in the PC-rich phase.<sup>3,24–27</sup>

For the partially miscible polymer blends, the  $\chi_{12}$  of the polymer blends can be determined by using eq. (5).<sup>3,24–27</sup>

$$\chi_{12} = \frac{\{(\phi_1'^2 - \phi_1''^2)[m_2 \ln(\phi_1''/\phi_1') + (m_1 - m_2)(\phi_2' - \phi_2'')] + (\phi_2'^2 - \phi_2''^2)[m_1 \ln(\phi_2''/\phi_2') + (m_2 - m_1)(\phi_1' - \phi_1'')]\}}{2m_1m_2(\phi_1'^2 - \phi_1''^2)(\phi_2'^2 - \phi_2''^2)} \quad (5)$$

where  $\phi_1'$  is the apparent volume fraction of polymer 1 dissolved in the polymer 1-rich phase,  $\phi_1''$  is the apparent volume fraction of polymer 1 in the polymer-2 rich phase.

The  $\chi_{12}$  values of the SPEEK–PC blends which are calculated from eq. (5) are presented in Table II. From Table II, the  $\chi_{12}$  values of the SPEEK–PC blends are found to range from 0.191

**Table II Polymer-Polymer Interaction Parameter ( $\chi_{12}$ ) of SPEEK-PEI, SPEEK-PC, and PEI-PC Blends**

Blend <sup>a</sup>	$\chi_{12}$		
	SPEEK-PEI <sup>b</sup>	SPEEK-PC <sup>c</sup>	PEI-PC <sup>d</sup>
0.2	-0.543	0.255	0.237
0.3	-0.487	0.272	0.246
0.4	-0.825	0.224	0.241
0.5	-0.303	0.236	0.224
0.6	-0.011	0.208	0.228
0.7	-0.200	0.191	0.235
0.8	-0.244	0.204	0.213

<sup>a</sup> Blend composition given as overall weight fraction SPEEK in SPEEK-PEI blend, SPEEK in SPEEK-PC blend, and PEI in PEI-PC blend.

<sup>b</sup> Data obtained from eq. (1).

<sup>c</sup> Data obtained from eq. (5).

<sup>d</sup> Data obtained from refs. 31 and 36.

to 0.272 with the blend composition. The  $\chi_{12}$  values of the PEI-PC blends are obtained from the earlier studies,<sup>31,36</sup> and found to range from 0.213 to 0.246 with the blend composition.

### Phase Behavior of SPEEK-PEI-PC Ternary Blends

Phase relationship for a ternary mixture where one component is a solvent have been first reported for the case of symmetric systems ( $\chi_{13} = \chi_{12}$ ) by Scott<sup>37</sup> and by Tompa<sup>38</sup> using the lattice theory of Flory and Huggins. For the mixtures of three monodisperse homopolymers, the spinodal for a ternary polymer blend is given by eq. (6).<sup>28</sup> For polydisperse polymers, it has been found that number-average molecular weights can be used in place of monodisperse molecular weights in the modified Flory-Huggins equation with very little effect on the consequent value of  $\chi$ .<sup>24,39</sup>

$$m_1\phi_1 + m_2\phi_2 + m_3\phi_3 - 2[m_1m_2(\chi_1 + \chi_2)\phi_1\phi_2 + m_2m_3(\chi_2 + \chi_3)\phi_2\phi_3 + m_3m_1(\chi_3 + \chi_1)\phi_3\phi_1] + 4m_1m_2m_3(\chi_1\chi_2 + \chi_2\chi_3 + \chi_3\chi_1)\phi_1\phi_2\phi_3 = 0 \quad (6)$$

where  $\chi_i = (\chi_{ij} + \chi_{ik} - \chi_{jk})/2$ ,  $m_1$ ,  $m_2$ , and  $m_3$  are the number of lattice of SPEEK, PEI, and PC, respectively. The  $m_1$ ,  $m_2$ , and  $m_3$  can be obtained by following relation:  $m_1 = V_1/V_0$ ,  $m_2 = V_2/V_0$ , and  $m_3 = V_3/V_0$  where  $V_1$ ,  $V_2$ ,  $V_3$ , and  $V_0$  are the molar volume of SPEEK, PEI, PC, and the repeating unit of PEI, respectively. The values of  $m_1 = 22.3$ ,  $m_2 = 20.3$ , and  $m_3 = 17.9$  were used for

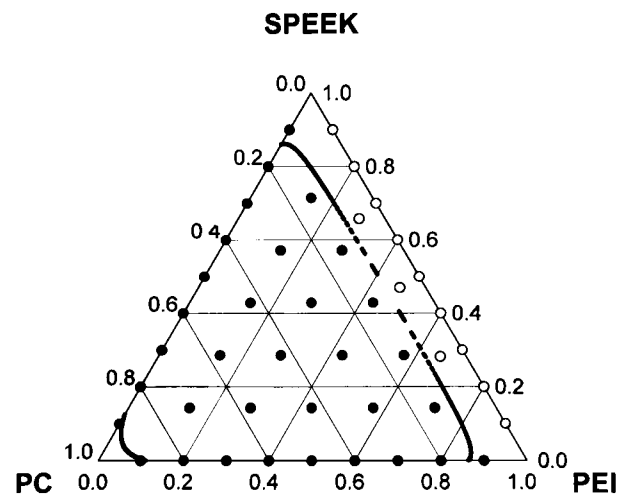
the SPEEK, PEI, and PC, respectively. A repeating unit of the PEI has been chosen as a site volume. The  $\chi_i$  in eq. (6) was obtained from the average value of  $\chi_{12}$  of the three binary blends.

The results of  $T_g$  of the SPEEK-PEI-PC ternary blends are shown in Figure 7. In Figure 7, the open circles indicate single  $T_g$  observation and the filled circles indicate two  $T_g$  observation by DSC. The spinodal curves of the SPEEK-PEI-PC ternary blends that were calculated from eq. (6) are also shown in Figure 7. In the SPEEK-PEI-PC ternary blends, phase separation regions that showed two  $T_g$  are found to be consistent with the spinodal curves calculated from the  $\chi_{12}$  values of the three binary blends.

### CONCLUSIONS

Binary blends of the SPEEK-PEI and SPEEK-PC, and ternary blends of the SPEEK-PEI-PC were investigated by DSC. From the results of the SPEEK-PEI blends by thermal analysis, single  $T_g$ s were observed at all the blend compositions. For the SPEEK-PC blends, double  $T_g$ s were observed. From these results, it is concluded that the blends of SPEEK-PEI is miscible and the blends of SPEEK-PC is immiscible.

Polymer-polymer interaction parameter ( $\chi_{12}$ ) of the SPEEK-PEI blends was calculated from the modified Lu and Weiss equation, and found to range from -0.011 to -0.825 with the blend composition. For the SPEEK-PC blends, the  $\chi_{12}$  val-



**Figure 7** Spinodal curves (- · - ·) and  $T_g$  of SPEEK-PEI-PC ternary blends: (○) single  $T_g$  and (●) double  $T_g$ .

ues were calculated from the modified Flory–Huggins equation, and found to range from 0.191 to 0.272 with the blend composition.

For the SPEEK–PEI–PC ternary blends, phase separation regions that showed two  $T_g$ s were found to be consistent with the spinodal curves calculated from the  $\chi_{12}$  values of the three binary blends.

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## REFERENCES

- Paul, D. R.; Newman, S. *Polymer Blends*, Academic Press: New York, 1978.
- Olabisi, O.; Robeson, L. M.; Shaw, M. T. *Polymer–Polymer Miscibility*, Academic Press: New York, 1979.
- Kim, W. N.; Burns, C. M. *Macromolecules* 1987, 20, 1876.
- Cowie, J. M. G.; Reilly, A. A. N. *Polymer* 1992, 33, 4814.
- Pugh, C.; Percec, V. *Macromolecules* 1986, 19, 65.
- Kim, H. I.; Pearce, E. M.; Kwei, T. K. *Macromolecules* 1989, 22, 3374.
- Ogawa, T.; Marvel, C. S. *J Polym Sci Polym Chem Ed* 1985, 23, 1231.
- Alomran, A.; Rose, J. B. *Polymer* 1996, 37, 1735.
- Kucera, F.; Jancar, J. *Polym Eng Sci* 1998, 38, 783.
- Trotta, F.; Drioli, E.; Moraglio, G.; Poma, E. B. *J Appl Polym Sci* 1998, 70, 477.
- Karcha, R. J.; Porter, R. S. *J Polym Sci Polym Phys Ed* 1993, 31, 821.
- Karcha, R. J.; Porter, R. S. *Polymer* 1992, 33, 4866.
- Lee, H. S.; Kim, W. N. *Polymer* 1997, 38, 2657.
- Chun, Y. S.; Lee, H. S.; Jung, H. C.; Kim, W. N. *J Appl Polym Sci* 1999, 72, 733.
- Nishi, T.; Wang, T. T. *Macromolecules* 1975, 8, 909.
- Harris, J. E.; Paul, D. R.; Barlow, J. W. *Polym Eng Sci* 1983, 23, 676.
- Masi, P.; Paul, D. R.; Barlow, J. W. *J Polym Sci Polym Phys Ed* 1982, 20, 15.
- DiPaola-Baranyi, G.; Fletcher, S. J.; Degre, P. *Macromolecules* 1982, 15, 885.
- Olabisi, O. *Macromolecules* 1975, 8, 316.
- Schmitt, B. J.; Kirste, R. G.; Jelenic, J. *Makromol Chem* 1980, 181, 1655.
- Schmitt, R. L.; Gardella, J. A., Jr.; Salvati, L., Jr. *Macromolecules* 1986, 19, 648.
- Lu, X.; Weiss, R. A. *Macromolecules* 1992, 25, 3242.
- Lu, X.; Weiss, R. A. *Macromolecules* 1991, 24, 4381.
- Kim, W. N.; Burns, C. M. *J Appl Polym Sci* 1987, 34, 945.
- Kim, W. N.; Burns, C. M. *Polym Eng Sci* 1988, 28, 1115.
- Kim, W. N.; Burns, C. M. *J Appl Polym Sci* 1990, 41, 1575.
- Lee, H. S.; Kim, W. N.; Burns, C. M. *J Appl Polym Sci* 1997, 64, 1301.
- Su, A. C.; Fried, J. R. *Polym Eng Sci* 1987, 27, 1657.
- Jin, X.; Bishop, T. M.; Ellis, S. T. *Br Polym J* 1985, 17, 4.
- Bishop, M. T.; Karasz, F. E. *Macromolecules* 1985, 18, 86.
- Chun, Y. S.; Lee, H. S.; Kim, W. N.; Oh, T. S. *Polym Eng Sci* 1996, 36, 2694.
- Christian, B.; Williams, D. J.; Karasz, F. E.; MacKnight, W. J. *Polymer* 1987, 28, 1009.
- Allard, D.; Prud'homme, R. E. *J Appl Polym Sci* 1982, 27, 559.
- Wang, L. F.; Pearce, E. M.; Kwei, T. K. *J Polym Sci Polym Phys Ed* 1991, 29, 619.
- Fox, T. G. *Bull Am Phys Soc* 1956, 1, 123.
- Chun, Y. S.; Lee, H. S.; Kim, W. N.; Oh, T. S. *Polymer (Korea)* 1995, 19, 913.
- Scott, R. L. *J Chem Phys* 1949, 17, 279.
- Tompa, H. *Trans Faraday Soc* 1949, 45, 1142.
- Narasimhan, V.; Huang, R. Y. M.; Burns, C. M. *J Polym Sci Polym Symp* 1986, 74, 265.