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Miscibility and synergistic effect of impact strength in polycarbonate/ABS blends

JONG-HAN CHUN
Chemical Safety Department, Industrial Safety Training Institute, Inchon, Korea
KI-SUCK MAENG
Department of Chemical Engineering, Chung Nam University, Dae Jeon, Korea
KWANG S. SUH
Department of Materials Science, Korea University, Seoul, Korea

Miscibility and mechanical properties of polycarbonate/acrylonitrile–butadiene–styrene (PC/ABS) blends prepared by a continuous kneader-type extruder were investigated. It was found that the PC/ABS blend consists of the miscible PC/SAN phases and immiscible PC/PB phases, which agrees well with that predicted by the group contribution method of solubility parameters. Differences in calculated solubility parameter ($\Delta\delta$) are 0.20 $(\text{cal}\,\text{cm}^{-3})^{1/2}$ for the PC/SAN pair and 1.85 $(\text{cal}\,\text{cm}^{-3})^{1/2}$ for the PC/PB pair. Interestingly, the impact strength of the present PC/ABS blends exhibits a synergistic effect over a much wider range of polycarbonate content, i.e. 20%–80%, than that of the previously reported ones. This may be attributed to the enhanced mixing and low butadiene content. However, further work on the relationship with the rubber particle size is required to understand fully this observation.

1. Introduction

PC/ABS blends have been widely used as one of the major engineering plastics. While PC has the highest notched impact strength in unmodified commercial engineering plastics, PC/ABS blends have an impact strength lower than PC itself. Because of this, much attention has been paid to the miscibility and impact properties of this blend system [1].

The impact strength of the polymer blends should depend strongly on whether or not the blends are miscible. One might expect that the impact strength will be higher in the miscible blend systems than in the immiscible blend systems by improving the interaction in the interfacial region between the components. However, the impact strength of the PC/ABS blends known to be miscible at all compositions shows a negative deviation from the additivity connecting the values for the pure PC and ABS. For example, Suarez *et al.* [2] employed a single screw extruder and obtained a negative deviation from the additivity for all compositions. On the other hand, Kurauchi and Ohta [3] employed a conventional twin screw extruder and obtained a synergistic effect, i.e. a positive deviation from the additivity, at 60%–90% PC content. These results suggest that if the blending equipment having a better mixing efficiency is employed, a synergistic effect at a wider range of PC content might be observed.

The aim of the present work, therefore, was to study the miscibility and the notched impact strength of the PC/ABS blends prepared by a continuous kneader-type extruder.

2. Experimental procedure

2.1. Materials and blending

The PC used in the study was Iupilon S-2000, injection moulding grade (Mitsubishi Gas Co., Japan) and ABS was supplied from Lucky Ltd, Korea. SAN with 26% acrylonitrile and 74% styrene, was selected because Keitz *et al.* [4] observed that the miscibility of PC and SAN was maximum at 25%–27% AN. Two ABSs, ABS-1 with 18% butadiene and ABS-2 with 40% butadiene, were prepared. Their final compositions are shown in Table I.

Blending of PC and ABS was done in a continuous kneader-type extruder, MDK/E (screw diameter 46 mm, screw length 506 mm). The processing temperature was set at 220–240 °C, which was selected to prevent the degradation which might occur above this temperature range. PC/ABS-1 blends were extensively used throughout the present study, because blending with ABS-2 produced a considerable backflow of the melt, possibly due to a high elasticity.

Test specimens for measuring such properties as the yield strength (ASTM D638, M-1 type) and Izod impact strength (ASTM D256, thickness 1/4 in., ~ 0.63 cm) were injection moulded under the conditions in Table II.

2.2. Mechanical and thermal properties

Yield strength was measured by Instron Universal Tensile Tester and the dynamic modulus was measured using a Rheometrics Dynamic Spectrometer

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TABLE I Monomer composition for the ABSs used in this study

Polymer	Acrylonitrile	Styrene	Butadiene
ABS-1	22	60	18
ABS-2	16	44	40

(RDS-7700, Rheometrics). Glass transition temperatures were obtained from the peaks in a loss modulus versus temperature plot by directly reading the peak maximum temperature. Differential scanning calorimetry (DSC) (1090, Du Pont) was also used to study the glass transition behaviour. Aluminium sample pans were used for all DSC experiments.

All measurements were carried out in a nitrogen atmosphere and at a heating rate of 10 °C min^{-1}. RDS was run at a frequency of 6.28 rad s^{-1}.

3. Results and discussion

3.1. Miscibility

Dynamic mechanical properties of the PC/ABS-1 blends were measured, and the results are shown in Fig. 1 for the storage modulus and in Fig. 2 for the loss modulus as a function of temperature. In these figures, as expected, a strong temperature dependence of dynamic modulus was observed. In Fig. 1, the storage modulus decreases sharply around 150 °C for the polycarbonate component, around 90 °C for the ABS-1 component, and between the two temperatures for the blends. At lower temperatures, differences in magnitudes of the storage modulus are observed. The results for the temperature dependence of the loss modulus indicate that the ABS component has two transitions, one near − 85 °C and the other near 105 °C. In addition, PC has one transition around 156 °C. These transitions, combined with the literature information, are assigned to the glass transitions of the butadiene phase, SAN phase, and PC phase, in order of decreasing temperature. For the peaks at a high-temperature region, a shift in peak temperatures can be observed. In contrast, the peak at a low-temperature region shows no change in peak temperatures regardless of the composition. Instead, some change in magnitude only can be seen. The value is the highest for ABS-1 and decreases as the PC content increases. The same trend was observed for the PC/ABS-2 blends, the typical data of which are given in Fig. 3. Glass transition temperatures were read directly from the temperatures where the maximum in a peak exists and tabulated in Table III.

A clear shift in the glass transition temperatures by changing the blend ratio can be seen in the DSC thermograms shown in Fig. 4. In this experiment, the

TABLE II Injection conditions for specimens used in this study

Injection conditions	PC	ABS	PC/ABS blends
Moulding temperature (°C)	100	60	60–90
Injection pressure (kg cm^{-2})	1200	900	900
Post injection pressure (kg cm^{-2})	500	500	500
Injection speed (mm s^{-1})	50	50	50
Cylinder temperature (°C)	280–300	190–230	200–290
Nozzle temperature (°C)	290	210	220–280
Preliminary drying	4 h at 120 °C	2 h at 70 °C	4 h at 70–100 °C

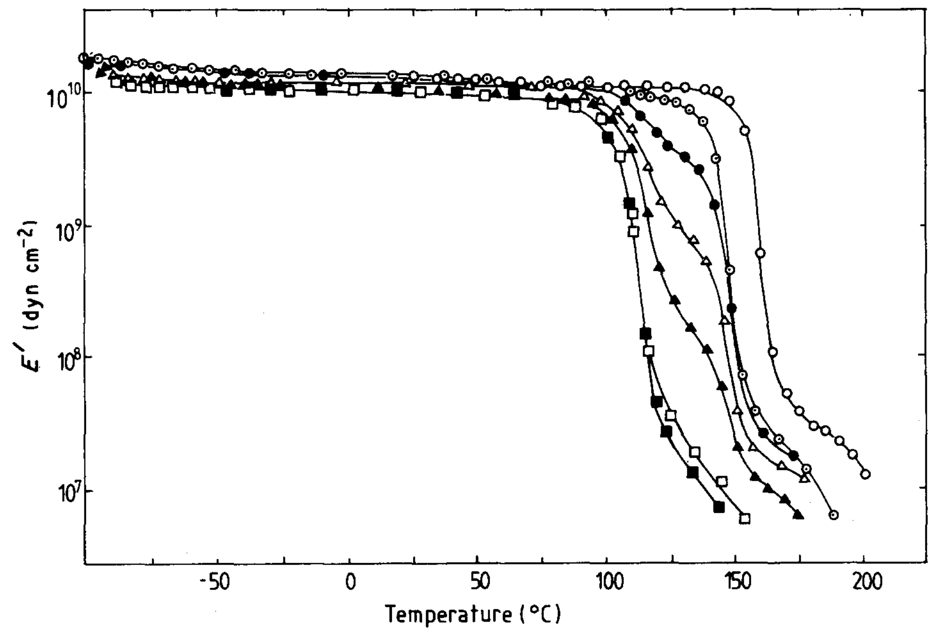

Figure 1 Storage modulus as a function of temperature for polycarbonate/ABS-1 blends. (○) PC, (⊙) PC/ABS = 90/10, (●) PC/ABS = 70/30, (△) PC/ABS = 50/50, (▲) PC/ABS = 30/70, (□) PC/ABS = 10/90, (■) ABS-1.

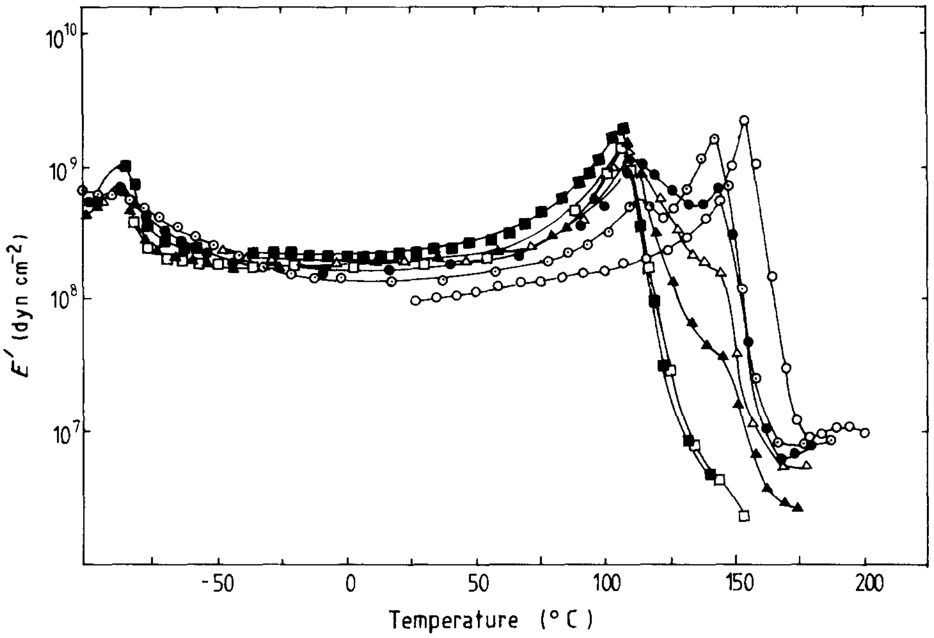

Figure 2 Loss modulus as a function of temperature for polycarbonate/ABS-1 blends. (○) PC, (⊙) PC/ABS = 90/10, (●) PC/ABS = 70/30, (△) PC/ABS = 50/50, (▲) PC/ABS = 30/70, (□) PC/ABS = 10/90, (■) ABS-1.

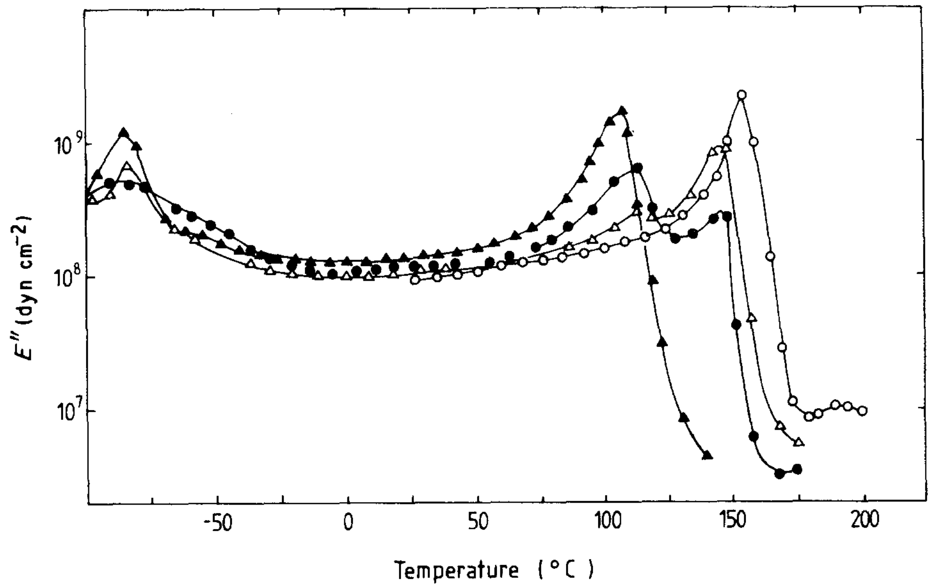

Figure 3 Loss modulus as a function of temperature for polycarbonate/ABS-2 blends. (○) PC, (●) PC/ABS = 90/10, (△) PC/ABS = 50/50, (▲) ABS-2.

TABLE III T_g for PC/ABS-1 blends estimated from the loss modulus curves

PC/ABS-1	$T_{g,b}$ (°C)	$T_{g,s}$ (°C)	$T_{g,p}$ (°C)
100/0			156
90/10	− 86	114	144
70/30	− 86	114	143
50/50	− 86	113	143
30/70	− 86	112	142
10/90	− 85	107	140
0/100	− 85	107	

Subscripts b, s and p represent butadiene, SAN, and PC phases, respectively.

peaks at the high-temperature region only were investigated because the peaks near − 85 °C showed no change in peak temperatures. For the temperature region investigated, the parent polymers, PC and ABS-1 show single transitions only, and the blends show two transitions.

A shift in transition temperatures is a well known phenomenon found in the miscible blend systems [5, 6]. The phases interact with each other at their interfaces so that the transition is affected, the net result being a shift in observed transition temperatures. Therefore, the present results suggest that, in the PC/ABS blends, the PC/SAN phases are miscible and the PC/PB phases are immiscible. In order to confirm the experimental observation, Small's group contribution theory [7] was employed to predict the miscibility of the PC/SAN and PC/PB phases in the PC/ABS blends. The following equation was used.

$$\delta = \frac{\rho \sum G}{M} \qquad (1)$$

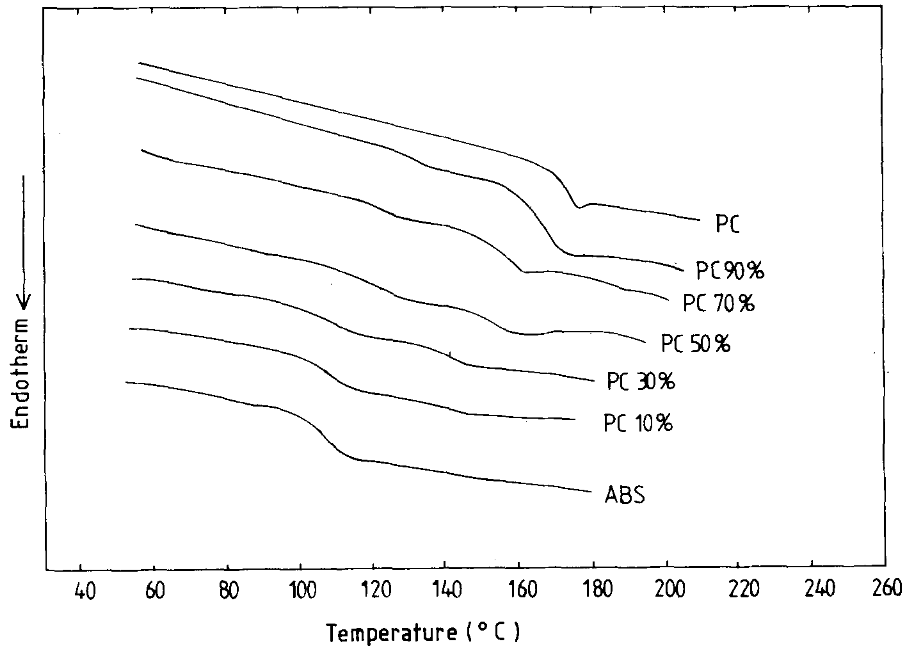

Figure 4 DSC thermograms for polycarbonate/ABS-1 blends.

where δ is the solubility parameter, ρ the density, G the group molar attraction constants and M the molecular weight. For a polymer, M is the mer molecular weight. In this calculation, solubility parameters of the PC/PB and PC/SAN pairs were estimated separately. The solubility parameters of PC and PB were calculated directly from the above equation. The solubility parameter of the SAN phase changes, depending on the concentration of the AN phase. For the present study, SAN with 26.8% AN was used, so that the solubility parameter was calculated from the interpolation method between those of PS and PAN. The calculated solubility parameters of PC, PB, PS, PAN, and SAN (26.8% AN) are 10.1, 8.25, 9.05, 12.4, and 9.90 $(\mathrm{cal\,cm^{-3}})^{1/2}$, respectively, which agree well with the published values [8–10]. From these values, the differences in the solubility parameters, $\Delta\delta$, are 0.20 $(\mathrm{cal\,cm^{-3}})^{1/2}$ for the PC/SAN pair and 1.85 $(\mathrm{cal\,cm^{-3}})^{1/2}$ for the PC/PB pair. This prediction, combined with Kozlowski and Skowronski's theory on the miscibility criteria [11] based on $\Delta\delta$, suggests that the PC/ABS blends are composed of two systems where one is miscible (PC/SAN phases) and the other immiscible (PC/PB phases).

3.2. Yield strength and impact strength

Yield strength and notched Izod impact strength of PC/ABS-1 blends as a function of temperature were measured using injection-moulded specimens; the results are shown in Fig. 5 for the yield strength and in Fig. 6 for the notched Izod impact strength. As shown in Fig. 5, the yield strength of the blends increases linearly with the PC content, which means that the PC/ABS-1 blends possess an additivity. Notched Izod impact strength, as shown in Fig. 6, does not seem to follow the additive principle. Izod impact strength in the intermediate composition region of roughly 20%–80% PC content shows a positive deviation from the additivity, i.e. a synergistic effect.

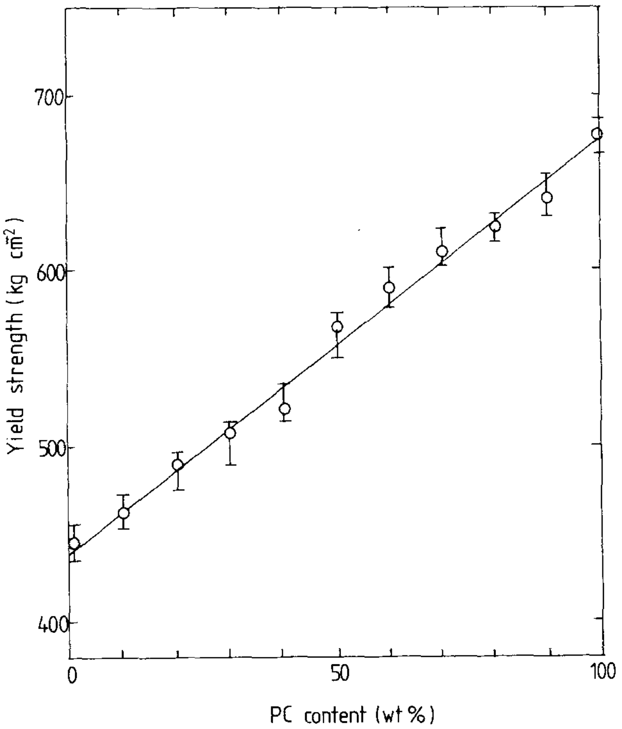

Figure 5 Yield strength of polycarbonate/ABS-1 blends as a function of polycarbonate content.

The present results differ, over a range of PC contents showing a synergistic effect, from the previously published results of Suarez *et al.* [2] and Kurauchi and Ohta [3]. These are compared in Fig. 7 and Table IV. The major differences in materials and blending equipments are summarized in Table IV. As shown in Fig. 7, Suarez *et al.* [2], using a single screw extruder, found no positive deviation from the additive line connecting the values for the pure PC and ABS. One of Kurauchi and Ohta's blends by the conventional twin screw extruder possesses a synergistic effect at 60%–90% PC content while the other does not [3]. On the other hand, the present blend

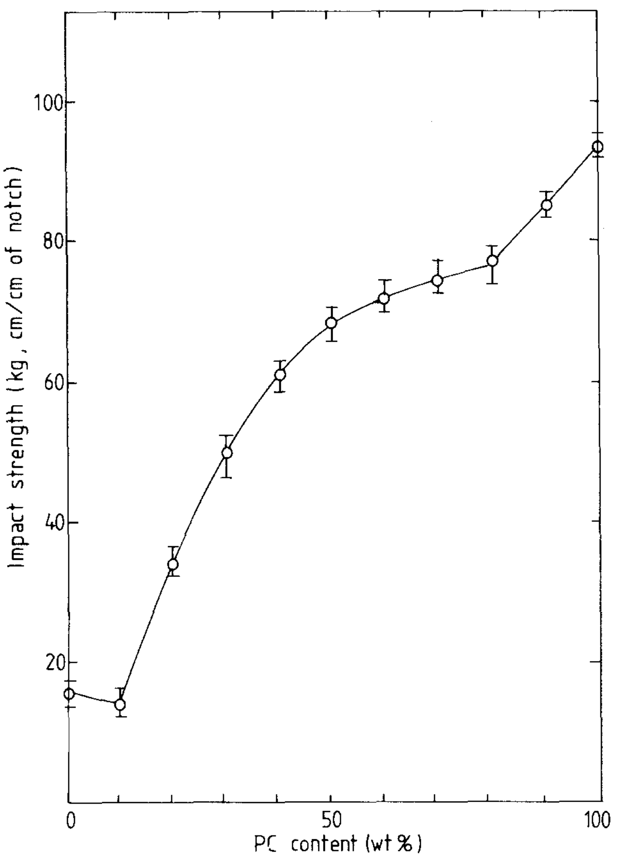

Figure 6 Notched Izod impact strength of polycarbonate/ABS-1 blends as a function of polycarbonate content.

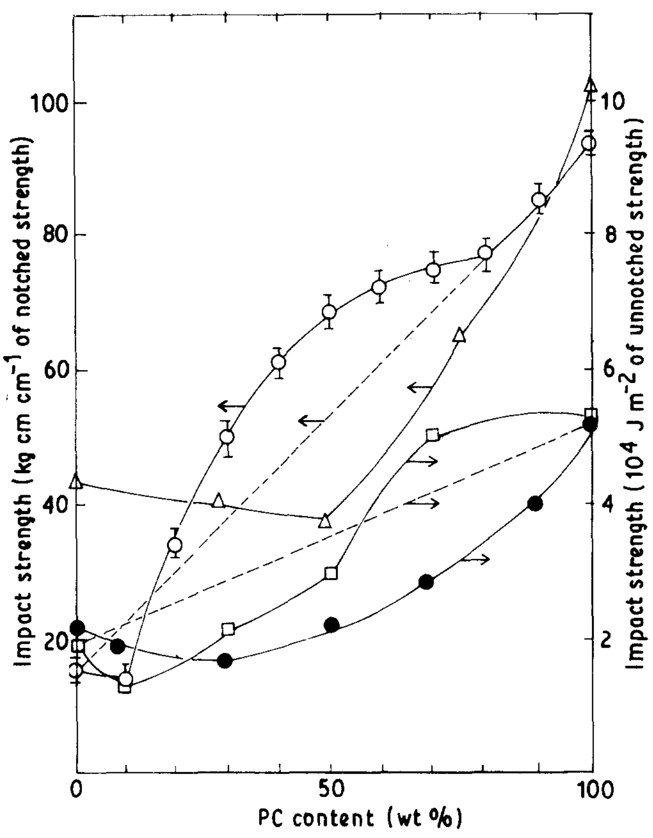

Figure 7 Comparison of impact strength from various works. (○) Present Work (PC/ABS-1), (△) [2], (□) [3] (PC/ABS of 29% PB), (●) [3] (PC/ABS of 37% PB), (---) additive rule.

system possesses a synergistic effect at 20%–80% PC content.

The differences between these works, as compared in Table IV, are the type of processing equipment and the content of butadiene phase. Apparently the range of PC content showing the synergistic effect has been broadened by using ABS with the lowest butadiene content (18%) and a continuous kneader-type extruder. These factors may be equally important in determining the impact strength of the PC/ABS blends. The impact strength of the rubber particle-dispersed blends is known to be governed primarily by the size of rubber particle and interparticle distance, which is again controllable by the type of equipment and butadiene content. In other words, with processing conditions fixed, an optimum butadiene content for an optimum size of rubber phase may exist and vice versa. Therefore, the exact reasons for such different observations between these works cannot be given. Work related to changes in rubber particle size and distribution is now in progress. Nevertheless, the present results indicate that the impact strength of the PC/ABS blends is controllable by both processing conditions and butadiene content in the ABS.

4. Conclusions

Study of the miscibility and mechanical properties of the PC/ABS blends prepared by a continuous kneader-type extruder has led to the following conclusions.

1. PC/SAN phases in the PC/ABS blends are miscible whereas PC/PB phases are not.
2. Small's group contribution method is useful in predicting the miscibility of each phase in the PC/ABS blends. Differences in the calculated solubility parameter ($\Delta\delta$) are 0.20 (cal cm^{-3})$^{1/2}$ for the PC/SAN phases and 1.85 (cal cm^{-3})$^{1/2}$ for PC/PB phases.
3. A synergistic effect on the impact strength of the PC/ABS blends at 20%–80% PC content, a much wider range of PC content than the previously reported ones, was observed. The reason for this, however, needs to be investigated further.

TABLE IV Comparison of various works on the PC/ABS blends

Reference	Acrylonitrile content (%)	Butadiene content (%)	Equipment	Synergistic effect (% PC)
[2]	–[b]	–[b]	Single screw	no S.E.[a]
[3]	20	37	Twin screw	no S.E.
	22	29		60–90
Present work	22	18	Continuous kneader	20–80
	16	40		–[b]

[a] S.E. = synergistic effect.

[b] No data available.

4. Impact strength of the PC/ABS blends may be improved by a careful design of processing conditions and material selection.

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