

Miscibility, Phase Behavior, and Mechanical Properties of Ternary Blends of Poly(vinyl chloride)/Polystyrene/Chlorinated Polyethylene-*graft*-Polystyrene

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ABSTRACT: The effectiveness of chlorinated polyethylene-*graft*-polystyrene (CPE-*g*-PS) as a polymeric compatibilizer for immiscible poly(vinyl chloride)/polystyrene (PVC/PS) blends was investigated. The miscibility, phase behavior, and mechanical properties were studied using differential scanning calorimetry (DSC), dynamic mechanical analysis (DMA), Izod impact tests, tensile tests, and scanning electron microscopy (SEM). DSC and DMA studies showed that PVC is immiscible with chlorinated polyethylene (CPE) in CPE-*g*-PS, whereas the PS homopolymer is miscible with PS in CPE-*g*-PS. The PVC/PS/CPE-*g*-PS ternary blends exhibit a three-phase structure: PVC phase, CPE phase, and PS phase that consisted of a PS homopolymer and PS in CPE-*g*-PS. The mechanical properties showed that CPE-*g*-PS interacts well with both PVC and PS and can be used as a polymeric compatibilizer for PVC/PS blends. CPE-*g*-PS can also be used as an impact modifier for both PVC and PS. SEM observations confirmed, after the addition of CPE-*g*-PS, improvement of the interfacial adhesion between the phases of the PVC/PS blends. © 1998 John Wiley & Sons, Inc. *J Appl Polym Sci* 69: 995–1003, 1998

Key words: PVC/PS blends; chlorinated polyethylene-*graft*-polystyrene; compatibilization; phase behavior; mechanical properties

INTRODUCTION

Blends of two polymers have been under intensive investigation in industrial and academic laboratories for many years because of the strong economic incentives.^{1–4} However, most polymer blends are immiscible and consist of two distinctive phases whose interfaces are weakly bounded with

poor interaction, and these result in poor mechanical properties of the blends. Some blends will produce materials with poor elongational characteristics, often referred to as cheesy, that must be upgraded before they can be considered for many end-use applications. It is usual to add a third polymer to an immiscible blend to increase the miscibility and/or to modify the interfacial adhesion of the blend and, hence, to improve the mechanical properties of the blends. The study of ternary blends becomes increasingly important^{5–11} with the incentive to discover new polymeric materials and/or to develop practical and economic methods for recycling commodity plastics.^{12–16}

In this article, we are concerned with the ternary blends of poly(vinyl chloride) (PVC)/poly-

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styrene (PS)/chlorinated polyethylene-*graft*-polystyrene (CPE-*g*-PS). It is well known that PVC/PS blends are immiscible and have rather poor mechanical properties for their poor interfacial adhesion. However, it has been reported by other authors^{17–20} that the mechanical properties of PVC/PS blends can be upgraded significantly by the addition of a third polymer, such as polystyrene-*block*-poly(methyl methacrylate) (PS-*b*-PMMA),¹⁷ the polystyrene-*block*-polycaprolactone (PS-*b*-PCL) block copolymer,¹⁷ and the epoxidized styrene-butadiene-styrene block copolymer (ESBS).¹⁸ Paul et al.^{19,20} used chlorinated polyethylene (CPE) as the third polymer and found that the properties are improved less dramatically because CPE only interacts with PVC and does not interact with PS at all. In our studies, CPE-*g*-PS was used as the third component. It was expected that the PS chains of CPE-*g*-PS would mix with the added PS homopolymer while the CPE of CPE-*g*-PS would interact with the PVC component through the polar-polar interaction.^{19,20} Thus, CPE-*g*-PS would serve as a compatibilizer to the immiscible PVC/PS blends. Our attention was paid to the effect of CPE-*g*-PS on the miscibility, phase behavior, and mechanical properties of PVC/PS blends. The techniques employed included differential scanning calorimetry (DSC), dynamic mechanical analysis (DMA), Izod impact tests, tensile tests, and scanning electron microscopy (SEM).

EXPERIMENTAL

Materials and Preparation of Blends

The PVC used in this study was PVC S-700, a commercial product of the Qilu Petrochemical Co. (Zipou, Shandong, China); it had an average degree of polymerization of 650–750. The PS (GPPS) with a melting index (MI) of 0.76 g/min (180°C, 5 kg) was obtained from the Jilin Chemical Industry Co. (Jilin, China). The CPE-*g*-PS with an MI = 0.77 g/min (180°C, 5 kg) was provided by the Anhui Research Institute of Chemical Technology (Hefei, Anhui, China). The CPE-*g*-PS was prepared by grafting styrene onto CPE, and the content of CPE in CPE-*g*-PS was 43.3 wt %. The CPE was a rubbery type with a 40 wt % chlorine content and was a commercial product of the Weifang Chemicals Factory (Shandong, China). Tribasic lead sulfate and dibasic lead stearate supplied by

the Wenzhou Plastics Additives Factory (Zhejiang, China) were used as thermal stabilizers for PVC. The glass transition temperature and the mechanical properties of the polymers used in this study are shown in Table I.

The PVC/PS blends without CPE-*g*-PS and added with 10 wt % CPE-*g*-PS were prepared by melt mixing. The blend compositions studied were 100/0, 80/20, 60/40, 40/60, 20/80, and 0/100 PVC/PS in terms of the weight ratio. The preweighed mixtures of PVC, PS, and CPE-*g*-PS together with stabilizers were first mixed in a high-speed mixer for 30 min at room temperature, then milled for 5 min on a two-roll mill at 160°C. The samples thus obtained were molded into sheets (about 4-mm thick) with a hot-press molding machine at 170°C and 100 atm for 5 min. To prevent possible degradation, 1.5 wt % of the stabilizers were added to the blends.

Differential Scanning Calorimetry

A Perkin-Elmer DSC-7 differential scanning calorimeter was employed to study the glass transition behavior of the samples. The instrument was calibrated with an indium standard, and the measurements were conducted under a nitrogen atmosphere. The sample weight used in the DSC cell was kept in the range of 10–15 mg; a heating rate of 20°C/min was used in all cases. The midpoint of the slope change of the heat-capacity plot of the second scan was taken as the glass transition temperature (T_g).

Dynamic Mechanical Measurements

Dynamic mechanical measurements were carried out on a Rheovibron Model DDV-III-EA dynamic viscoelastometer (Toyo Baldwin Co., Japan). The frequency used was 35 Hz and the heating rate was 3.0°C/min. The specimen dimension was 6.0 × 0.4 × 0.2 cm³.

Izod Impact Tests

Notched Izod impact tests were done on an AFS/MK3-654S/000 Izod impact tester (CEAST Co., Germany) at 20°C. The ASTM D256 standard was used, and a minimum of five specimens with a notch angle of 45° were tested in each case.

Tensile Tests

Tensile tests were carried out on an AG-5000A testing machine at 20°C. Standard dumbbell spec-

Table I Glass Transition Temperature and Mechanical Properties of the Polymers Used in This Study

Polymer	T_g (by DSC) (°C)	IS (J/m)	E (GPa)	σ_y (MPa)	σ_b (MPa)	ϵ_b (%)
PVC	82.7	31.8	2.37	56.5	47.8	125
PS	100.3	13.3	2.41	—	31.1	5.8
CPE- <i>g</i> -PS	-2.6, 105.4	109.8	1.87	17.1	15.1	38.7

All the data were obtained from this study. IS, Izod impact strength; E , Young's modulus; σ_y , stress at yield; σ_b , stress at break; ϵ_b , elongation at break.

imens (ASTM D638) with a $2.5 \times 0.6 \times 0.4 \text{ cm}^3$ neck were used. A minimum of five specimens were tested in each case. The crosshead speed was 5 mm/min, corresponding to a relative strain rate of 0.2 min^{-1} .

Morphological Observation

To observe the fracture surfaces of PVC/PS blends and PVC/PS/CPE-*g*-PS blends, the specimens were fractured under cryogenic conditions using liquid nitrogen. A Hitachi X-60 scanning electron microscope (SEM) was used for observation, before which the surfaces were coated with thin layers of gold of 200 Å.

RESULTS AND DISCUSSION

Phase Behavior

Figures 1 and 2 show the DSC thermograms (the second scan) of the PVC/PS blends without CPE-*g*-PS and with 10 wt % CPE-*g*-PS, respectively, and the results are summarized in Tables II and III, respectively. It can be seen from Figure 1 that the PVC/PS blends without CPE-*g*-PS exhibit two separate T_g 's, one at about 83°C and the other at about 101°C, which correspond to the glass transition temperature of PVC and PS, respectively. These two T_g 's are composition-independent, indicating the immiscibility of PVC and PS. By the addition of 10 wt % CPE-*g*-PS, the T_g of the PVC phase in the blends remains constant and is composition-independent, whereas the T_g of the PS phase in the blends slightly shifts to high temperature (Fig. 2). Except for the PVC/CPE-*g*-PS blend in which the T_g of the PS phase (105.8°C) is close to that in the CPE-*g*-PS copolymer (105.4°C), the T_g of the PS phase in the other PVC/PS/CPE-*g*-PS blends is also composition-independent and is located at about 103°C, which is between the T_g of the PS homopolymer

(100.3°C) and that of the PS in the copolymer (105.4°C). For all the PVC/PS/CPE-*g*-PS blends, there exists another transition which is located at about -5 to 0°C and is due to the glass transition of CPE in CPE-*g*-PS. The above results suggest that PVC is immiscible with CPE in CPE-*g*-PS, whereas the PS homopolymer is miscible with PS in the CPE-*g*-PS copolymer.

All the binary and the ternary blends were also analyzed using DMA. The loss modulus (E'') versus T curves of the PVC/PS blends without CPE-*g*-PS and with 10 wt % CPE-*g*-PS are shown in Figures 3 and 4, respectively. The T_g data ob-

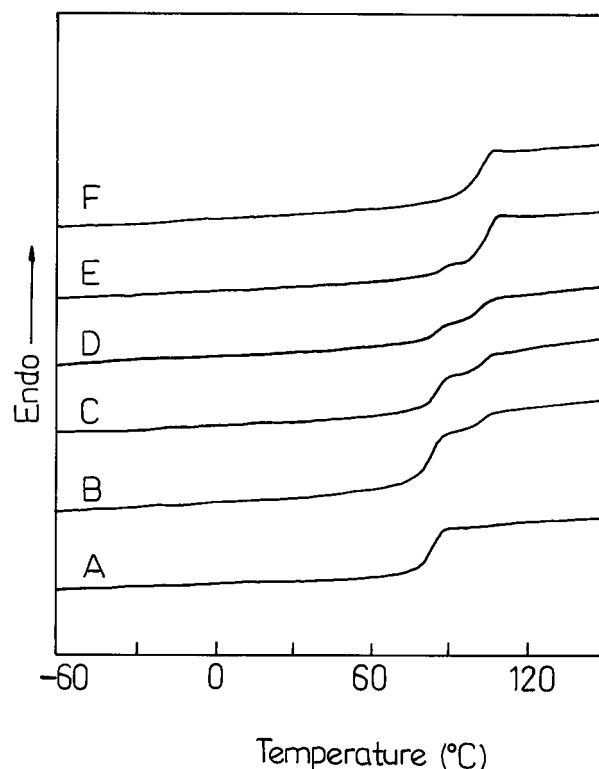


Figure 1 DSC thermograms of the second scans of PVC/PS binary blends. PVC/PS: (A) 100/0; (B) 80/20; (C) 60/40; (D) 40/60; (E) 20/80; (F) 0/100.

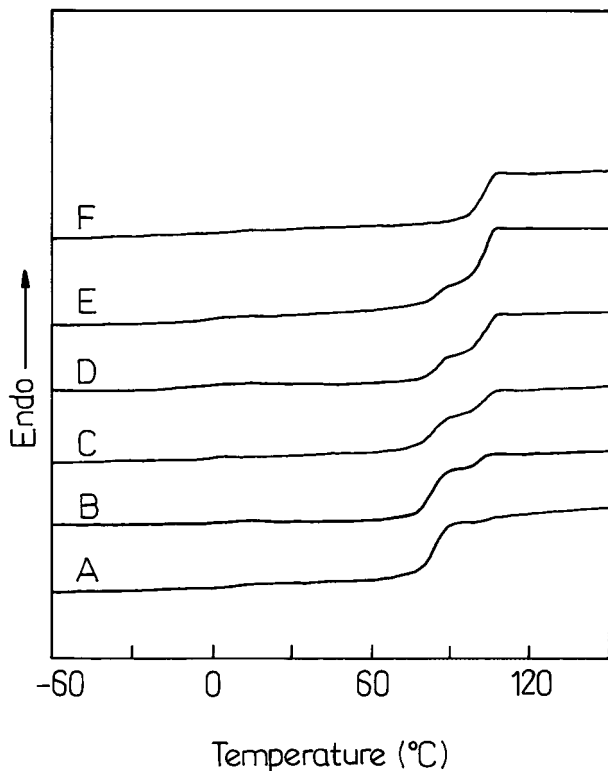


Figure 2 DSC thermograms of the second scans of PVC/PS/CPE-*g*-PS ternary blends. PVC/PS: (A) 100/0; (B) 80/20; (C) 60/40; (D) 40/60; (E) 20/80; (F) 0/100.

tained from Figures 3 and 4 are also listed in Tables II and III, respectively. For the PVC/PS blends without CPE-*g*-PS, two relaxation peaks appear above 60°C (Fig. 3). The higher is located at about 131°C, which is due to the glass transition of PS, and the strength of this relaxation peak decreases with decrease of the PS content in the blends. The lower is located at about 111°C, which is due to the glass transition of PVC, and its

strength increases with increase of the PVC content in the blends. The positions of these two relaxation peaks are both composition-independent, suggesting the immiscibility of PVC and PS. In the range of -50 to 0°C, a broad relaxation peak is shown, which is due to the secondary transition of PVC and its strength decreases with decrease of the PVC content in the blends. For the PVC/PS/CPE-*g*-PS blends, the E'' versus T curves are essentially the same as those of the PVC/PS blends without CPE-*g*-PS except for the following (Fig. 4): (1) For all the PVC/PS/CPE-*g*-PS blends, the curves show a distinct relaxation peak at about 10°C, which is due to the glass transition of CPE in CPE-*g*-PS. (2) For the PVC/CPE-*g*-PS blend, a shoulder exists at about 139°C, which is due to the glass transition of PS in the copolymer. (3) The relaxation peaks of PS in the PVC/PS/CPE-*g*-PS ternary blends slightly shift to higher temperature compared to those in the blends without CPE-*g*-PS. The results presented here confirm the conclusion of the DSC study: PVC is immiscible with CPE in CPE-*g*-PS, whereas the PS homopolymer is miscible with PS in the copolymer. Therefore, the PVC/PS/CPE-*g*-PS blends exhibit a three-phase structure: the PVC phase, the CPE phase, and the PS phase that consists of the PS homopolymer and PS chains in the CPE-*g*-PS copolymer.

It should be pointed out that it is still possible that there exists some interaction between PVC and CPE in spite of their immiscibility. Because both PVC and CPE are polar chlorine-containing polymers, the interaction may result from the polar-polar interaction.^{19,20}

Impact Strength

Incorporation of PS in PVC results in a considerable decrease in the impact strength over that of

Table II Glass Transition Temperature of PVC/PS Blends Without CPE-*g*-PS

PVC/PS	T_g (°C) (by DSC)		T_g (°C) (by DMA)	
	PVC Phase	PS Phase	PVC Phase	PS Phase
100/0	82.7	—	110.0	—
80/20	82.0	101.9	111.0	131.5
60/40	82.9	101.0	111.1	129.0
40/60	82.9	101.7	112.0	132.0
20/80	83.6	100.5	111.8	131.0
0/100	—	100.3	—	130.1

Table III Glass Transition Temperature of PVC/PS Blends with 10 wt % CPE-*g*-PS

PVC/PS	T_g (°C) (by DSC)			T_g (°C) (by DMA)		
	PVC Phase	PS Phase	CPE Phase	PVC Phase	PS Phase	CPE Phase
100/0	83.9	105.8	-1.0	110.5	138.9	10.1
80/20	83.5	102.7	-3.4	111.1	133.9	10.1
60/40	83.2	103.0	-2.0	111.5	133.1	9.5
40/60	83.2	103.8	-4.8	112.0	134.1	10.1
20/80	83.6	103.0	-3.3	111.1	133.2	10.1
0/100	—	101.8	-2.5	—	134.5	9.0

pure PVC. From Figure 5, it can be seen that the impact strength of PVC/PS blends without CPE-*g*-PS decreases monotonically with increase of the PS content: The impact strength decreases greatly with the PS content up to 20 wt % and then continues to decrease slightly with further increase of the PS content. This can be attributed to the immiscibility and the poor interfacial adhesion between the PVC phase and the PS phase.

However, the variation of the impact strength of the PVC/PS blends with 10 wt % CPE-*g*-PS against the blend composition (also in Fig. 5) is complicated. Compared with the value of the

PVC/PS blends without CPE-*g*-PS, the impact strength of the PVC/PS/CPE-*g*-PS blend is higher at the compositions of 100/0, 60/40, 20/80, and 0/100 PVC/PS. The increase of the impact strength of the 100/0 and 0/100 PVC/PS blends, that is, the pure component polymers, after adding 10 wt % CPE-*g*-PS can be due to the toughening of PVC or PS by CPE-*g*-PS. CPE-*g*-PS consists of two phases, that is, the CPE rubbery phase and the PS glassy phase, and it has a good impact property (Table I). When PVC or PS is blended with CPE-*g*-PS, the CPE rubbery phase was well dispersed in the continuous PVC or PS phase, and

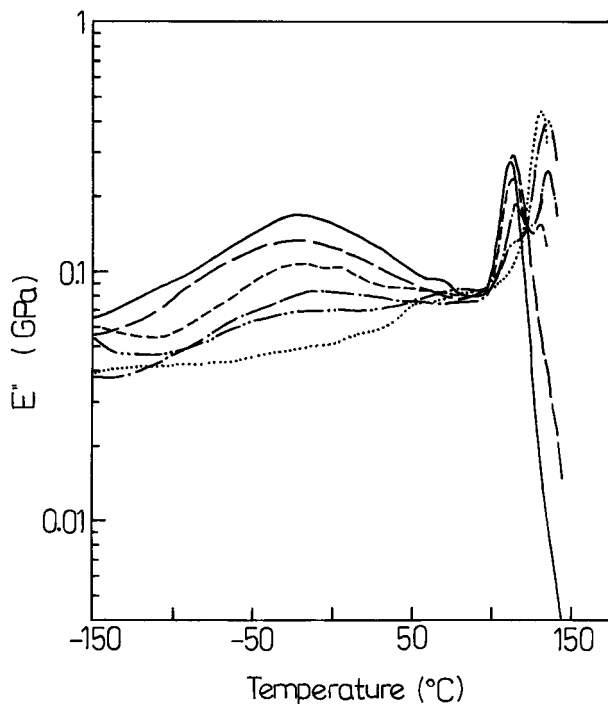


Figure 3 Temperature dependence of E'' of PVC/PS binary blends with compositions (—) 100/0, (— —) 80/20, (— — —) 60/40, (— · — ·) 40/60, (— · · — ·) 20/80, and (· · · · ·) 0/100 PVC/PS.

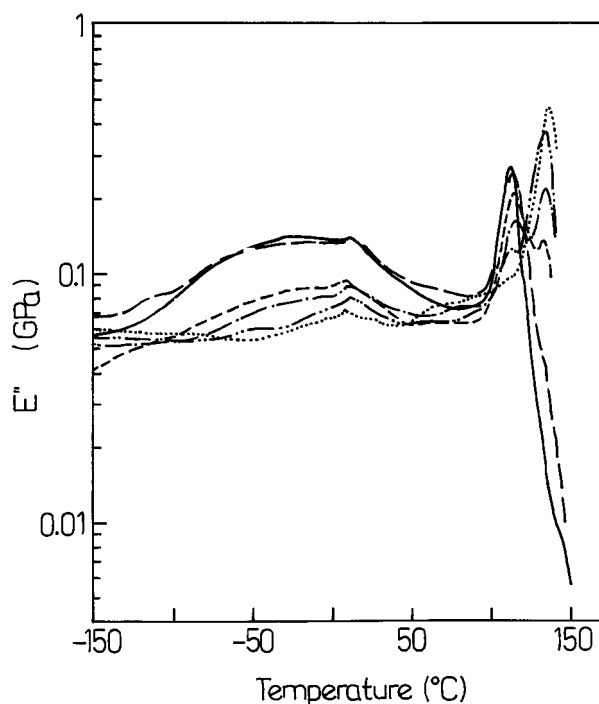


Figure 4 Temperature dependence of E'' of PVC/PS/CPE-*g*-PS blends with compositions (—) 100/0, (— —) 80/20, (— — —) 60/40, (— · — ·) 40/60, (— · · — ·) 20/80, and (· · · · ·) 0/100 PVC/PS.

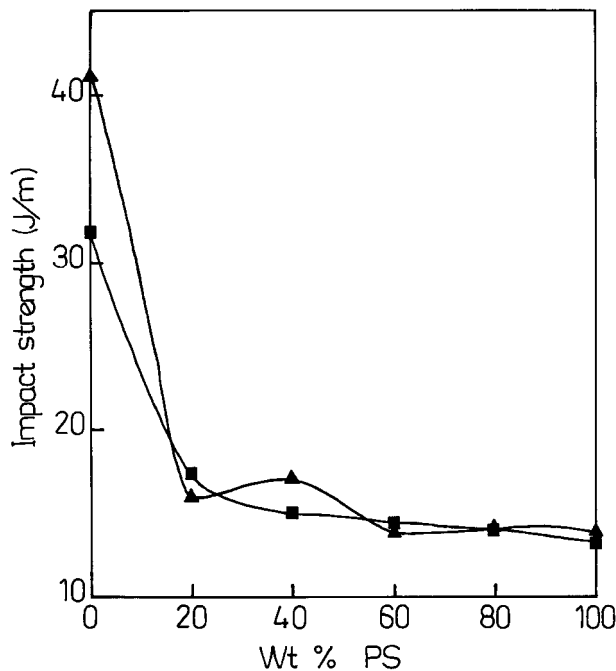


Figure 5 Composition dependence of notched Izod impact strength (20°C) of PVC/PS blends: (■) without CPE-*g*-PS; (▲) added with 10 wt % CPE-*g*-PS.

the interfaces between CPE and the continuous phase are well bounded. For this reason, the introduction of CPE-*g*-PS causes the improvement of the impact strength of PVC and PS.

At the same weight ratio of PVC to the PS homopolymer, the relative content of PS in the PVC/PS/CPE-*g*-PS blend is higher than that in the PVC/PS blend without CPE-*g*-PS and this results in a decrease of the impact strength because of the rather poor impact property of PS compared with PVC. However, CPE-*g*-PS can act as a compatibilizer because it interacts rather well with both PVC and PS. The interfacial activity of CPE-*g*-PS is dependent on the distribution of CPE-*g*-PS on the interfaces between the PVC phase and the PS phase. The larger the interfacial area between the PVC phase and the PS phase, the more CPE-*g*-PS there is on the interfaces and the more effectively the interfacial adhesion improves and, hence, the impact strength improves. The competition of the reduction in the impact strength caused by increase of the relative content of PS and the enlargement in the impact strength caused by the improved interfacial adhesion results in decrease of the impact strength of 80/20 and 40/60 PVC/PS blends and increase of the impact strength of the 60/40 PVC/PS blend after addition of CPE-*g*-PS. For the 20/80 PVC/PS

blend, the slight increase of the impact strength may be concerned with the toughening by brittle particles.²¹⁻²³

Tensile Properties

The common stress-strain behavior was obtained for both PVC/PS blends without CPE-*g*-PS and with 10 wt % CPE-*g*-PS. The stress-strain curves of the pure PVC and the PVC/CPE-*g*-PS blend exhibited characteristics of a ductile fracture, that is, obvious yield and neck formation were observed. However, for the pure PS and the other blends, no obvious yield was observed on the stress-strain curves, which shows that these materials are basically brittle at the strain rate of 0.2 min⁻¹ and room temperature (20°C). From the initial slopes, the Young's moduli of PVC, PS, and all the blends were calculated. In Figure 6, the Young's moduli of the PVC/PS blends without CPE-*g*-PS and with 10 wt % CPE-*g*-PS are plotted as a function of blend composition. It can be seen that the Young's modulus of the PVC/PS blends is much lower than that of either of the pure components and shows a minimum at a 60/40 PVC/PS composition. Marked negative deviations from simple additivity are observed. These are typical of an essentially immiscible system.^{24,25} After ad-

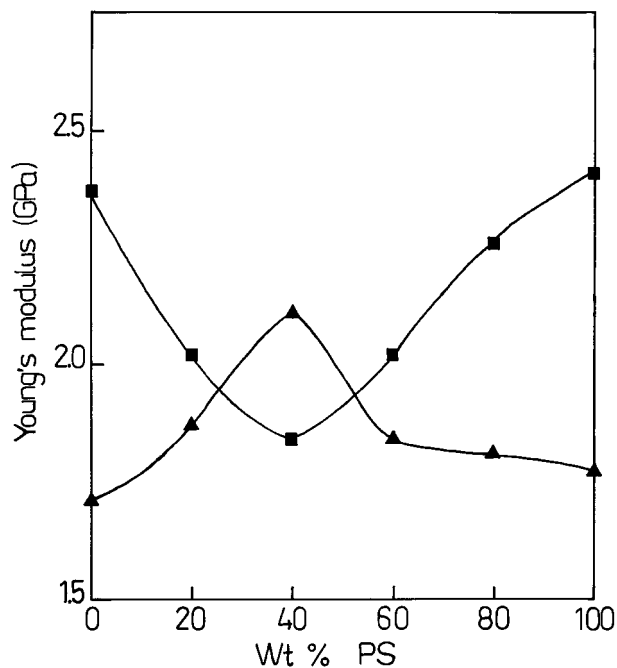


Figure 6 Composition dependence of Young's modulus (20°C) of PVC/PS blends: (■) without CPE-*g*-PS; (▲) added with 10 wt % CPE-*g*-PS.

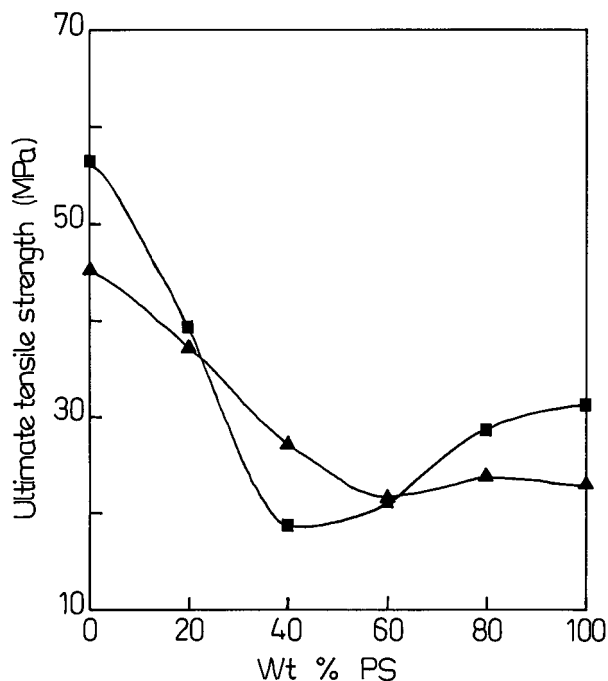


Figure 7 Composition dependence of ultimate tensile strength (20°C) of PVC/PS blends: (■) without CPE-*g*-PS; (▲) added with 10 wt % CPE-*g*-PS.

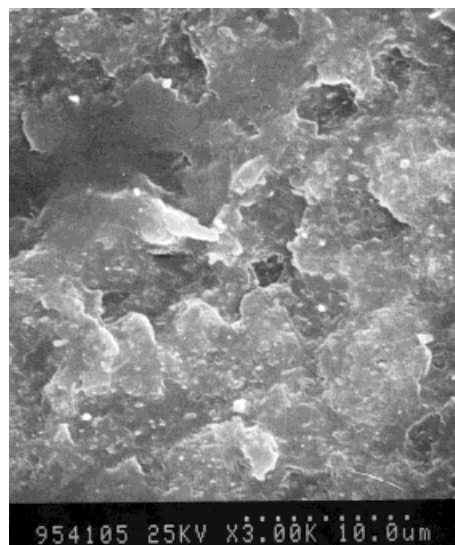
dition of CPE-*g*-PS, the Young's modulus shows positive deviations from simple additivity over the entire composition range, which can be attributed to the improvement of interfacial adhesion because CPE-*g*-PS interacts rather well with both PS and PVC. It is interesting to note that, after adding CPE-*g*-PS, the Young's modulus is substantially enhanced at a composition of 60/40 PVC/PS; however, the Young's modulus cannot be improved by the other compositions. In fact, with these compositions, the Young's modulus for the blends added with CPE-*g*-PS is lower than those for pure PVC/PS blends. These may be due to the very poor tensile properties of pure CPE-*g*-PS, which totally deteriorated the mechanical properties of the blends (Table I).

From stress-strain curves, we also obtained the ultimate tensile strengths of both PVC/PS blends without CPE-*g*-PS and with 10 wt % CPE-*g*-PS, and their variations against the blend composition are shown in Figure 7. For the PVC/PS blend without CPE-*g*-PS, at compositions ranging from 80/20 to 20/80 PVC/PS, the ultimate tensile strength is much lower than the corresponding weight-average value and exhibits marked negative deviations from simple additivity. After addition of 10 wt % CPE-*g*-PS, the negative deviations of the ultimate tensile strength from simple addi-

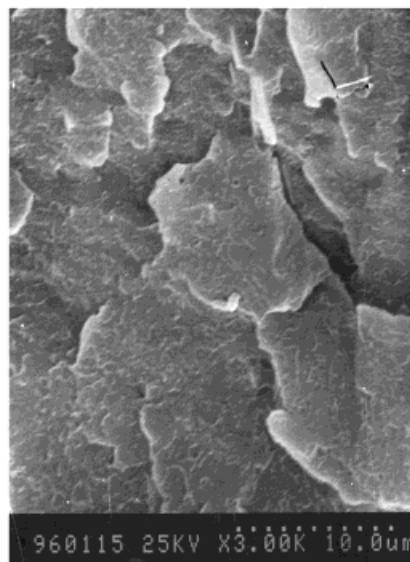
tivity becomes unmarked and, especially, the ultimate tensile strength of the 60/40 and 40/60 PVC/PS blends are improved. All these are consistent with the results of the Young's modulus and are due to the improvement of the interfacial adhesion between the PVC phase and the PS phase after adding CPE-*g*-PS.

Morphology

The morphology of the PVC/PS blends without CPE-*g*-PS and with 10 wt % CPE-*g*-PS were in-



(a)



(b)

Figure 8 Scanning electron micrographs of fractured surfaces of (a) PVC/CPE-*g*-PS and (b) PS/CPE-*g*-PS blends.

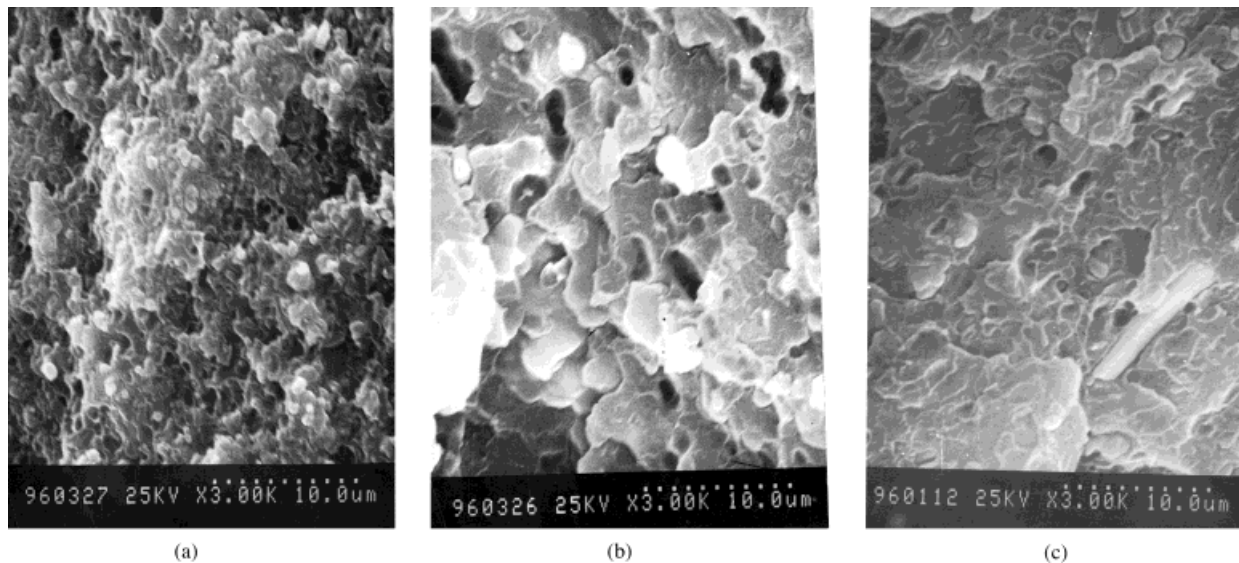


Figure 9 Scanning electron micrographs of fractured surfaces of PVC/PS binary blends. PVC/PS: (a) 60/40; (b) 40/60; (c) 20/80.

investigated using a scanning electron microscope (SEM). The specimens were fractured under cryogenic conditions using liquid nitrogen. The SEM photographs of the 90/10 PVC/CPE-*g*-PS blend and the 90/10 PS/CPE-*g*-PS blend are shown in Figure 8. Figure 8(a,b) clearly shows that the CPE domains were dispersed on a fine scale in the PVC and PS continuous phases, respectively. The average domain size is less than 1 μm in diameter. It can also be seen from Figure 8 that the

interfacial adhesion between the phases in the blends is rather good. These results confirm that there exist interactions between PVC and CPE in CPE-*g*-PS as well as between PS and CPE-*g*-PS. Figures 9 and 10 show SEM photographs of the PVC/PS binary blends and the PVC/PS/CPE-*g*-PS ternary blends, respectively. It can be seen from Figure 9 that the PVC/PS binary blends clearly exhibit a two-phase structure. The dispersed phase is pulled out of its cavities, leaving

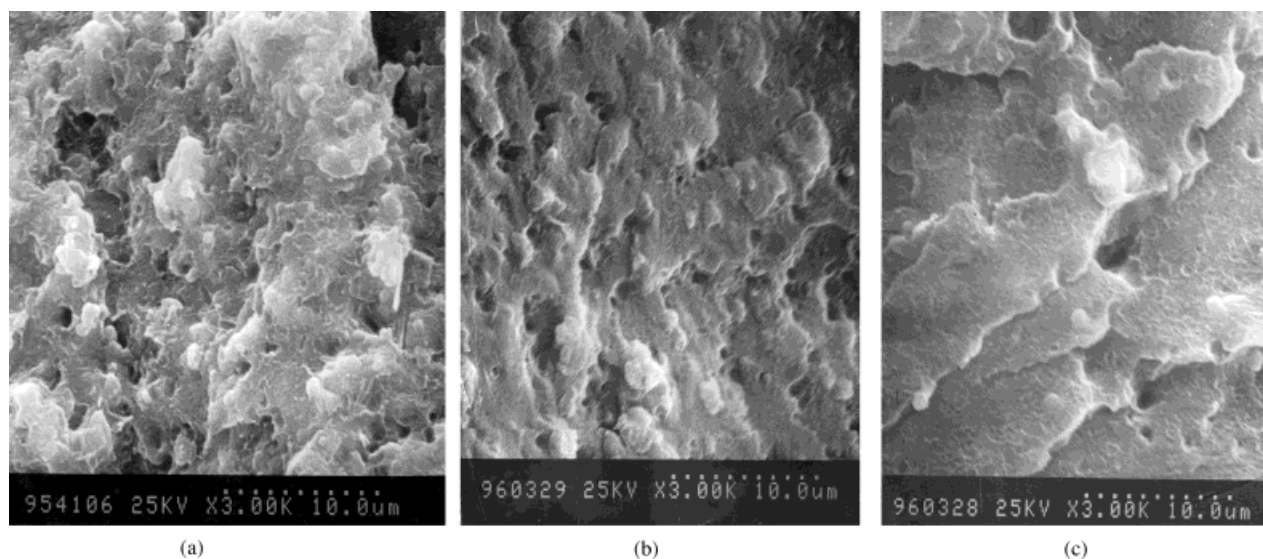


Figure 10 Scanning electron micrographs of fractured surfaces of PVC/PS/CPE-*g*-PS ternary blends. PVC/PS: (a) 60/40; (b) 40/60; (c) 20/80.

holes with a smooth surface. The interfaces between PVC and PS are weakly bonded, and the pure PVC/PS blends exhibit a coarse dispersion and a poor adhesion. It can also be seen from Figure 9 that fracture occurred in a macroscopically brittle manner. After adding of 10 wt % CPE-*g*-PS, it can be seen from Figure 10 that the morphology is modified, and the interfaces between phases become unclear and the interfacial adhesion is rather good as well. It is also noted that the PVC/PS blends to which CPE-*g*-PS was added even exhibit some characteristics of ductility (Fig. 10), although the fractures are macroscopically brittle. The morphological observations of the blends are consistent with the results of the impact strength and tensile properties.

CONCLUSIONS

In conclusion, this work has shown that the PS homopolymer is miscible with the PS chains in CPE-*g*-PS whereas PVC is immiscible with CPE in CPE-*g*-PS. There exist interactions between CPE-*g*-PS and PVC as well as between CPE-*g*-PS and PS, and CPE-*g*-PS can be used as a compatibilizer for the immiscible PVC/PS blends. CPE-*g*-PS can also be used as an impact modifier for both PVC and PS. SEM observations showed the improvement of the interfacial adhesion between the phases of the PVC/PS blends after the addition of CPE-*g*-PS.

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