

Polycarbonate/Acrylonitrile–styrene–acrylic elastomer Terpolymer Blends with Enhanced Interfacial Adhesion and Surface Gloss

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ABSTRACT: A blend of bisphenol A polycarbonate (PC) and an acrylonitrile–styrene–acrylic elastomer (ASA) terpolymer with high surface gloss and excellent interfacial properties was developed for automobile applications. Because PC and the styrene-*co*-acrylonitrile (SAN) copolymer that formed the matrix in the PC/ASA blend were not miscible, two different types of compatibilizers were examined to improve the compatibility of the blend. A diblock copolymer composed of tetramethyl polycarbonate and poly(methyl methacrylate) (PMMA) was more effective than PMMA in increasing interfacial adhesion between PC and

SAN. The surface gloss of the PC/ASA blend was always lower than that of the pure ASA included in the blend because of PC existing at the surface of the injection-molding specimen. The PC/ASA blend with optimum surface gloss and enhanced interfacial adhesion was developed through the control of the molecular weight of PC and the compatibilizer. © 2005 Wiley Periodicals, Inc. *J Appl Polym Sci* 96: 2097–2104, 2005

Key words: blends; interfaces; polycarbonates; surfaces

INTRODUCTION

Bisphenol A polycarbonate (PC) is used in a wide variety of applications because of its excellent balance of properties.¹ However, deficient characteristics of PC deter its application in some areas. A number of approaches have been used to enhance its poor characteristics, such as the thickness dependence of the notched impact strength and the poor radiation, solvent, and hydrolysis resistance. Blends of PC with acrylonitrile–butadiene–styrene (ABS) materials provide products with an improved balance of properties at reduced cost in comparison with PC. For these reasons, blends of PC with ABS materials have been commercially available for many years.^{1–11} However, these blends have two main drawbacks. In a typical PC/ABS blend, all rubber particles within the styrene-*co*-acrylonitrile (SAN) copolymer form a dispersed phase in the matrix composed of PC and SAN. Because PC blends with SAN are not miscible, the interfacial adhesion is not strong enough. The application

of blends with phase-separated structures is often limited because of their poor adhesion at the weld.^{11–15} Other problems stem from the physical (or chemical) aging of butadiene rubber. Butadiene rubber, containing a double bond in its repeat unit, undergoes physical (or chemical) aging caused by ultraviolet (UV) radiation in outdoor applications. The aging of butadiene rubber results in a continuous decline in the mechanical strength and color changes. Improvements in the interfacial adhesion and the reduction of aging are essential for broadening the applications of PC/ABS blends.

In this work, interfacial agents for PC/ABS blends were examined to enhance interfacial adhesion between PC and SAN, and then acrylic rubber, that is, poly(butyl acrylate), was added to prevent the aging problems of butadiene rubber. In previous studies,^{14,15} we explored block copolymers, such as PC-*b*-PMMA [where PMMA is poly(methyl methacrylate)], PC-*b*-SAN, TMPC-*b*-PMMA (where TMPC is tetramethyl polycarbonate), and TMPC-*b*-SAN, as interfacial agents for PC/SAN blends. In this study, PMMA was also explored as an interfacial agent for PC/SAN blends, and the results were compared with those of the TMPC-*b*-PMMA block copolymer, which was the most efficient compatibilizer of the block copolymers examined previously.¹⁵ PC does not form a miscible blend with PMMA when both components have high molecular weights. However, PC and PMMA do form

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TABLE I
Polymers Used in this Study

Polymer	Copolymer composition ^a	M_w^a	M_w/M_n^a	Rubber content ^a	Surface gloss ^b	η_0 (Pa s)	Source
PC-1	—	32,400	2.06	—	—	4,990	LG Chemicals (200-15)
PC-2	—	47,000	1.82	—	—	8,500	LG Chemicals (300-15)
ASA-1	24% AN	—	—	30 wt%	89	—	LG Chemicals (ASA 919)
ASA-2	24% AN	—	—	30 wt%	94	—	LG Chemicals (ASA 927)
ASA-3	24% AN	—	—	30 wt%	95	—	LG Chemicals (ASA 925)
SAN 24	24% AN	174,500	2.21	—	—	950	LG Chemicals (SAN 80HF)
PMMA	—	103,000	1.83	—	—	—	LG Chemicals (HI 830)
TMPC- <i>b</i> -PMMA ^c	—	88,300	—	—	—	—	Synthesized

M_w , weight-average molecular weight; M_n , number-average molecular weight.

^a Information was provided by the supplier.

^b Surface gloss was measured by ASTM D 523-89.

^c M_w of TMPC = 23,000.

a miscible blend of the molecular weight of one or both components is reduced because of the small positive interaction energy, that is, 0.03 cal/cm³.^{11,16–20} This means that blends of PC with PMMA are not miscible but appear to be right on the edge of miscibility. PMMA also forms miscible blends with SAN containing 9–33 wt % acrylonitrile (AN).^{21–24} These results indicate that PMMA is a potential interfacial agent for PC/SAN blends. Various acrylonitrile–styrene–acrylic elastomer (ASA) terpolymers, instead of ABS, were examined to improve the UV stability of the blends. Even though ASA has excellent UV stability, the surface gloss of a PC/ASA blend is generally lower than that of a PC/ABS blend. The surface gloss of ASA can be controlled by changes in the rubber particle size and distribution, rubber shape, and AN content of the SAN matrix.²⁵ The surface gloss of ASA can be enhanced by an increase in the AN content of SAN. However, the AN content of the SAN copolymer was fixed at approximately 24 wt % because the PC/SAN blend exhibited maximum interfacial adhesion at this AN concentration.^{14,15} To enhance the surface gloss of the blend, we controlled the particle size of the rubber, the rubber shape, the viscosity ratio of PC to ASA, and the type of compatibilizer and its concentration.

EXPERIMENTAL

The polymers used in the experiments and relevant information about them are listed in Table I. Polycarbonates (PC-1 and PC-2), ASA (ASA-1, ASA-2, and ASA-3), SAN, and PMMA were provided by LG Chemicals (Taejon, Korea). TMPC-*b*-PMMA copoly-

mers were synthesized in the laboratory. The procedure for the synthesis of TMPC-*b*-PMMA has been described in detail elsewhere.^{14,15} Blends of PC with the SAN copolymer were prepared via melt mixing or solution casting. PC/SAN mixtures were mixed in a Haake Rheomix torque rheometer (Karlsruhe, Germany) for 15 min at 60 rpm and 240°C. The blend films were also prepared via solution casting from dichloromethane in a petri dish. The solvent was allowed to evaporate slowly at room temperature, and the formed films were dried in a vacuum oven at 90°C for a week. Blends of PC and ASA were also prepared via melt mixing in a twin extruder at 240°C.

The interfacial tension between PC and SAN, or that between PC and SAN containing various amounts of a compatibilizer, was measured with an embedded-fiber-retraction technique.^{26–29} Fibers of PC were produced with an Instron fiber-spinning apparatus (Canton, MA). SAN copolymer sheets 0.15 mm thick, containing various amounts of a compatibilizer, were prepared through compression molding at 220°C. PC fibers and SAN plates were dried *in vacuo* at 170 and 110°C, respectively, for 24 h to ensure that no elastic effects related to the fiber orientation occurred. PC fibers were then placed between two plates of the SAN copolymer. The samples were subsequently transferred to a microscope equipped with a hot stage and a temperature controller. The samples were annealed at 180°C for 2 h before the fiber-retraction process was observed with an image analyzer at 240°C. The zero-shear viscosities (η_0) of PC and the SAN copolymer were measured with a Rheometrics RDS-II dynamic spectrometer (Piscataway, NJ) over a shear-rate range of 10⁻¹–10³ rad s⁻¹ under a nitrogen atmosphere at

240°C. The η_0 values obtained from the low-frequency plateau viscosities are listed in Table I.

The interfacial fracture toughness was measured with an asymmetric double-cantilever beam geometry.^{30–32} The polymers were compression-molded into rectangular plates (5.0 cm × 1.0 cm × 0.2 cm with a chrome-plated mold). Films of SAN copolymers containing various amounts of the block copolymer were also obtained via solution casting from dichloromethane. The samples were joined together in their respective molds at 200°C for 2 h under a slight pressure. The samples were allowed to cool in the mold for 3 h until they reached room temperature; this suppressed the formation of thermal stress. To examine the compatibilization effects, we also prepared PC and SAN sheets covered with thin films of the SAN copolymer containing a compatibilizer under the same conditions. Because the elastic moduli and crazing stresses of PC and SAN were different, the asymmetric double-cantilever beam test geometry was chosen to avoid crack propagation toward the more compliant material^{30–32} (if the more compliant material has a lower crazing stress, then crazes will grow at an angle away from the interface). An asymmetry was induced by the attachment of an SAN rectangular bar, with a lower crazing stress than that of the PC rectangular bar, to a rigid substrate (in this case, a 5-mm glass plate). The specimens were dried in a vacuum oven at 100°C for a day before the test. The fracture toughness was measured by the insertion of a single-edge razor blade at the interface. The wedge was pushed at a constant velocity of 4 $\mu\text{m/s}$ with a stepping motor. The crack length ahead of the wedge was measured with an optical microscope after crack propagation was allowed for 24 h.

The number-average diameter of the dispersed particles in the blend was measured with a Bummi Universe Co. I-Top digital image analyzer (Seoul, Korea). The specimens, prepared with a torque rheometer, were mounted on a Linkam THMS 600 hot stage equipped with a Linkam TMS 92 temperature controller (Waterfield, UK). The samples were heated rapidly to 240°C, and then changes in the size of the dispersed particles were monitored for a day. Because changes in the average diameter were not observed for that day, the samples annealed at 240°C for 30 min were used to measure the average diameter of the dispersed particles. The diameters of all the particles included in 10 separate photographs were measured, and the mean average was reported as the average diameter of the dispersed particles.

The mechanical and thermal properties of the PC/ASA blends were measured according to the ASTM method. To examine the weld-line strength of the blends, we prepared by injection molding specimens having the same shape as the tensile test bar and containing a weld line at the center. The tensile

strength of these sample bars was defined as the weld-line strength. The blend morphology was observed with scanning electron microscopy (SEM; JSM-840A, JEOL, Tokyo, Japan) and transmission electron microscopy (TEM; JEM-2000EX II, JEOL). To obtain image contrast for the SEM observations, we etched out the PC-rich phase in the blend samples by immersing the samples in an aqueous solution containing 30 wt % NaOH. The samples were stained with RuO_4 and then microtomed for TEM observations.

RESULTS AND DISCUSSION

Interfacial tension and adhesion

It is well recognized that the interactions between PC and SAN copolymers containing various amounts of AN are not favorable enough to produce complete miscibility, although evidence for various extents of miscibility has been reported.^{11,14–20} Among the various PC/SAN blends, it is also known that a PC blend with SAN containing about 24 wt % AN (SAN 24) shows optimum interfacial adhesion, regardless of the blend compositions. Because of this, the experiments reported hereafter were performed with SAN 24.

The interfacial tension is an important factor in determining the morphology of an immiscible blend. The embedded-fiber-retraction technique was developed as a dynamic method for measuring the interfacial tension between molten, high-molecular-weight polymers.^{26–29} The standard equilibrium methods have limitations in measuring interfacial tension between highly viscous polymers because of factors such as prohibitively long equilibrium times and the thermal degradation of polymers. The embedded-fiber-retraction technique involves the microscopic tracking of the shape evolution of a short embedded fiber and uses interfacial tension as a driving force for the retraction process. As shown in eq. (1),^{26–29} the slope of the function-related retraction shape of the embedded fiber $\{\lambda[f(R/R_0) - f(R_e/R_0)]\}$ versus the retraction time (t_r) yields the interfacial tension (γ):

$$\lambda[f(R/R_0) - f(R_e/R_0)] = t_r \gamma \quad (1)$$

where R is the effective radius of the retraction fiber, R_e is its value at time $t = 0$, and R_0 is the radius of a sphere with a value equal to the volume of the fiber. The function $f(R/R_0)$ and λ in eq. (1) are given by

$$f(x) = \frac{3}{2} \ln \left(\frac{\sqrt{1+x+x^2}}{1-x} \right) + \frac{3^{1.5}}{2} \arctan \left(\sqrt{3} \frac{x}{2+x} \right) - \frac{x}{2} - \frac{4}{x^2} \quad (2)$$

$$\lambda = (\eta_m + 1.7\eta_f)R/2.7 \quad (3)$$

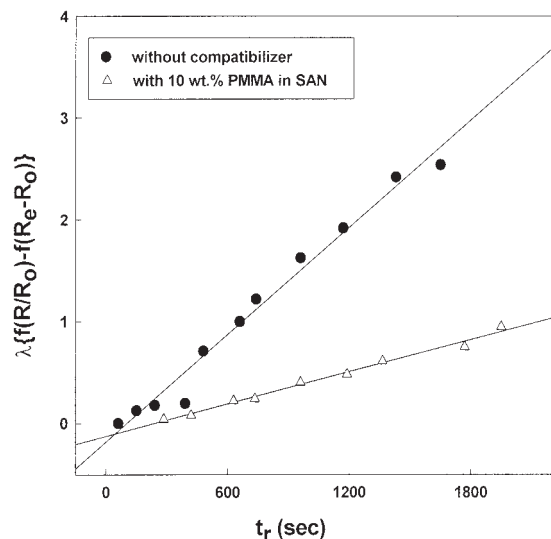


Figure 1 Plot of $\lambda [f(R/R_0) - f(R_e/R_0)]$ versus t_r for a PC fiber.

where x is equal to R/R_0 or R_e/R_0 and η_m and η_f are the zero-shear bulk viscosities of the matrix and fiber, respectively. As predicted in eq. (1), the experimental data shown in Figure 1 do yield a straight line. To explore the effect of the addition of the compatibilizer, we prepared SAN matrices containing various amounts of compatibilizer. The changes in the interfacial tension of the PC/SAN 24 blend as a function of the compatibilizer content are shown in Figure 2. The interfacial tension decreased very rapidly for compatibilizer concentrations of 0–10 wt % and then leveled

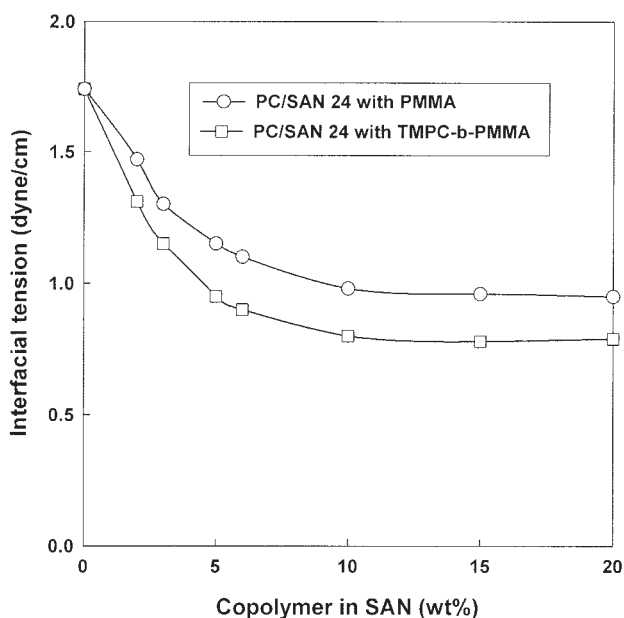


Figure 2 Effects of the compatibilizer content on the interfacial tension between PC and SAN 24.

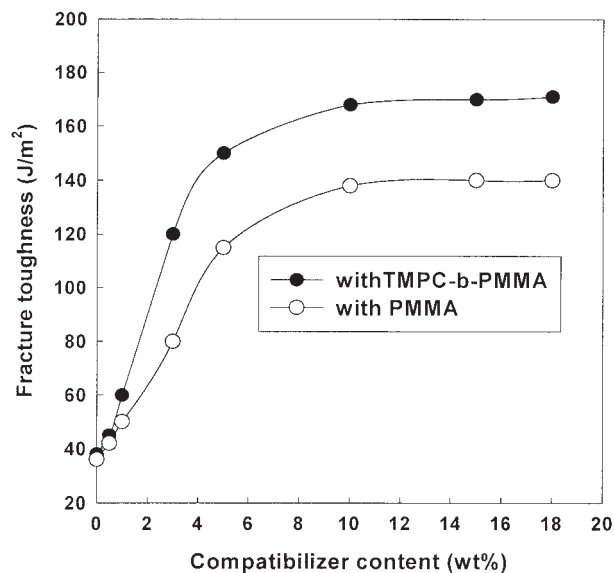


Figure 3 Fracture toughness of the PC/SAN 24 adhesive joints containing various amounts of the compatibilizer.

off. The TMPC-*b*-PMMA block copolymer was more effective than PMMA in reducing interfacial tension.

As described elsewhere for the asymmetric double-cantilever beam fracture test,^{30–32} the adhesion energy (G_c) was obtained from the crack length measured with a traveling microscope:

$$G_c = \frac{3u^2 E_{PC} D_{PC}^3}{8a^3 [1 + 0.64(D/a)]^4} \quad (4)$$

where u and a are the thickness of the wedge and the crack length, respectively. E_{PC} (2300 MPa) and D_{PC} are the elastic modulus and thickness of PC, respectively. The changes in the fracture toughness were examined through changes in the compatibilizer content of the SAN 24 films, which were placed between PC and SAN sheets for the compression molding. The fracture toughness of the PC/SAN 24 blend increased with increasing TMPC-*b*-PMMA content, with values as high as 170 J/m², and then approached an asymptotic value, as shown in Figure 3.

Dispersed particle and weld-line strength

The minor component in an immiscible blend forms droplets, that is, domains dispersed in the continuous phase mainly composed of the major component of the blend. To determine the optimum content of a compatibilizer for a blend, we examined changes in the average diameter of the dispersed droplets with the compatibilizer content. The changes in the average diameter of the dispersed particles in the PC/SAN 24 (7/3) blend are shown in Figure 4 as a function of the compatibilizer content. The average diameter de-

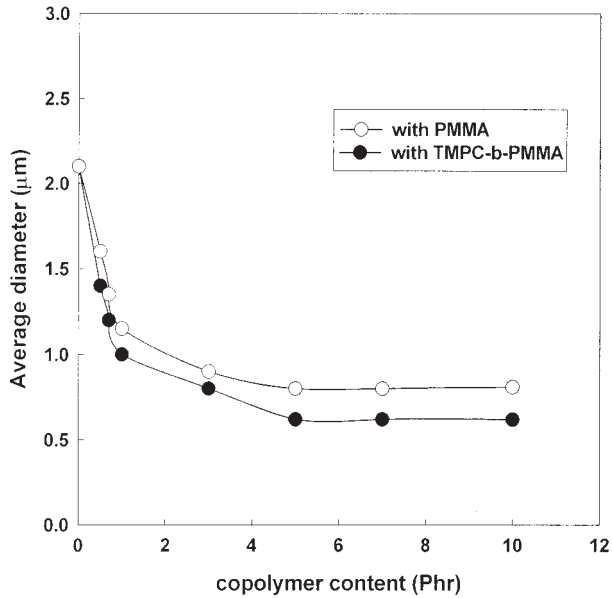


Figure 4 Average diameter of the dispersed particles for the 7/3 PC/SAN 24 blends as a function of the block copolymer content.

creased for the blends with 0–5.0 phr (parts per hundred parts of resin) compatibilizer and then leveled off at a fixed size. TMPC-*b*-PMMA was more effective than PMMA in reducing the average diameter of the dispersed particles in the PC/SAN blends. A similar trend was observed for the other compositions. The average diameter of the dispersed particles in the PC/SAN 24 (7/3) blends was further reduced from 2.1 to

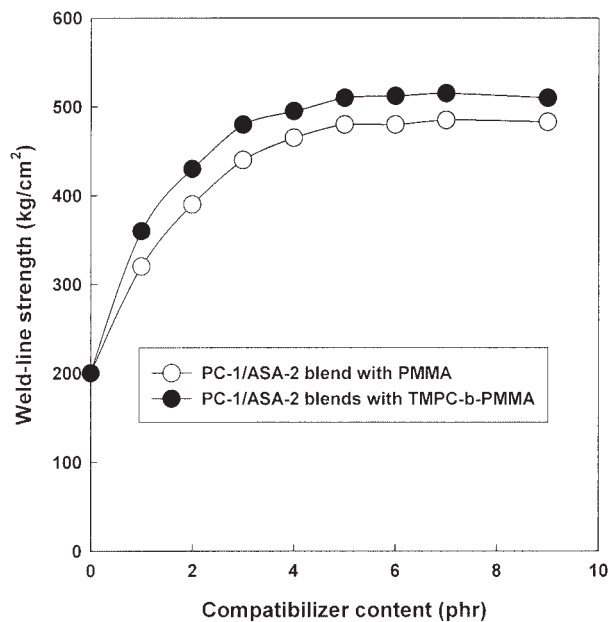
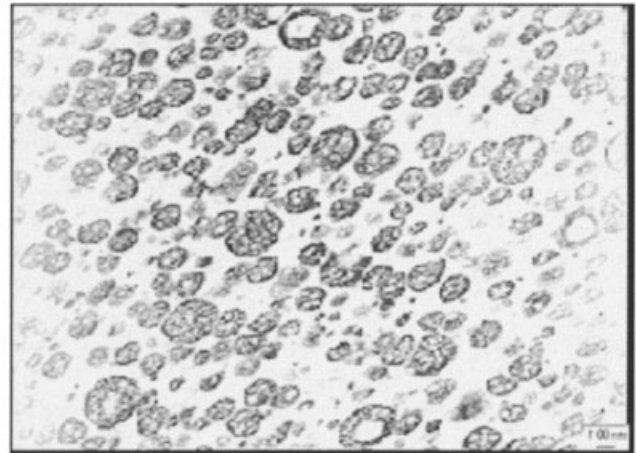
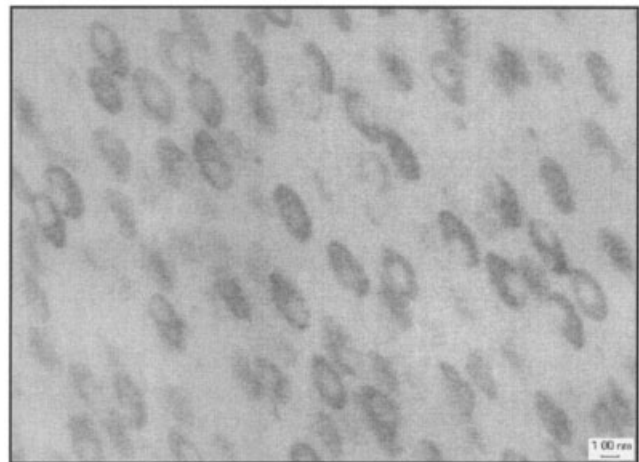


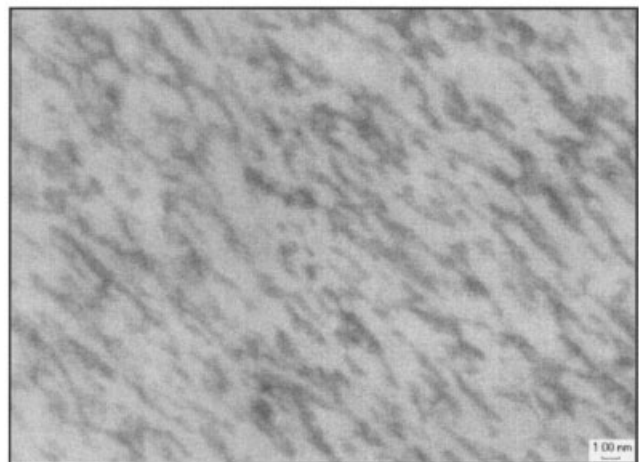
Figure 5 Weld-line strength of PC/SAN 24 blends as a function of the compatibilizer content.



(a)



(b)



(c)

Figure 6 TEM microphotographs of ASA terpolymers: (a) ASA-1, (b) ASA-2, and (c) ASA-3.

TABLE II
Mechanical and Thermal Properties of PC/ASA Blends

Property	Test method	PC-1/ASA-1/ PMMA	PC-1/ASA-2/ TMPC- <i>b</i> - PMMA	PC-1/ASA-2/ PMMA	PC-1/ASA-3/ PMMA	PC-2/ASA-2/ PMMA
Tensile strength (kg/cm ²)	ASTM D 638	600	590	590	585	610
Tensile elongation (%)	ASTM D 638	120	150	150	70	155
Flexural strength (kg/cm ²)	ASTM D 790	910	900	890	900	900
Flexural modulus (kg/cm ²)	ASTM D 790	23,700	24,000	23,500	24,000	24,000
Izod impact strength (25°C, kg cm/cm)	ASTM D 256	67	70	70	55	72
Izod impact strength (-10°C, kg cm/cm)	ASTM D 256	10	30	29	17	30
HDT (1/4 265 kg/cm ² , °C)	ASTM D 648	106	106	105	105	107
Surface gloss of the blend	ASTM D 523-89	87	89	91	92	94
Surface gloss of ASA	ASTM D 523-89	89	94	94	95	94

6/4 PC/ASA blends containing 5 phr compatibilizer were examined. HDT, heat distortion temperature.

0.7 μm by the addition of the TMPC-*b*-PMMA copolymer.

The weld-line strength of the specimens containing a weld line at the center of the tensile test bar was examined according to ASTM D 638. Figure 5 shows the weld-line strength of 6/4 PC-1/ASA-2 blends containing various amounts of compatibilizers. The weld-line strength increased for the blends containing 0–5.0 phr compatibilizer and then leveled off at a fixed value, as shown in Figure 5. TMPC-*b*-PMMA was more effective than PMMA in increasing the weld-line strength of the blends. For these reasons, the experimental results reported hereafter are those for the blends containing 5 phr compatibilizer.

Mechanical properties and surface gloss of the PC/ASA blends

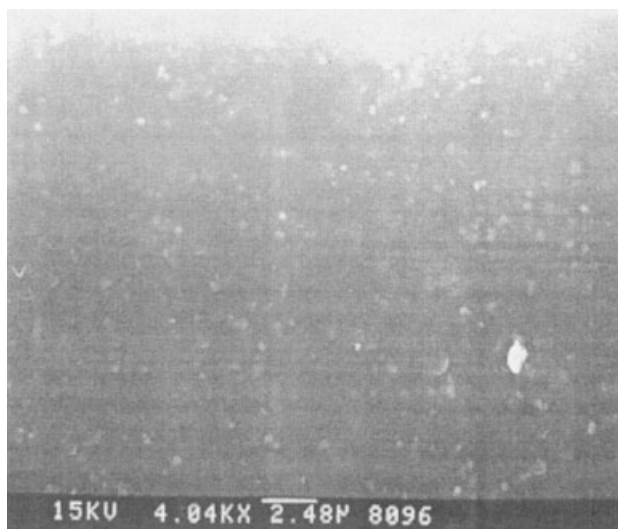
The surface gloss of the ASA terpolymer depends on the rubber particle size, particle size distribution, and shape of the rubber particles.²⁵ Here, three different types of ASA terpolymers were examined. As shown in Figure 6, the average diameters of ASA-1 and ASA-2 were the same (0.4 μm), although the rubber particles in ASA-1 contained SAN within the rubber phase. The surface gloss of ASA-2 was higher than that of ASA-1, as listed in Table I. ASA-3 (average diameter of rubber particles = 0.1 μm) exhibited the best surface gloss of the ASA terpolymers examined here.

Blends containing 60 wt % PC-1, 40 wt % ASA, and 5 phr compatibilizer were prepared, and then their mechanical properties and surface gloss were examined, as shown in Table II. All the blends exhibited similar mechanical properties, except for the impact

strength. The surface gloss of the blends was in the following order: PC-1/ASA-3 > PC-1/ASA-2 > PC-1/ASA-1. This means that the surface gloss of the PC/ASA blend depended on that of ASA. As shown in Table II, the blend including ASA-1 exhibited the lowest impact strength at a low temperature (-10°C). Because rubber particles including SAN within the rubber phase always deteriorated with respect to the impact strength at low temperatures and the surface gloss, they were not considered for further experiments. The surface gloss of the blends slightly increased with decreasing rubber particle size. However, a decrease in the rubber particle size was not desirable because of a decline in the impact strength, as shown in Table II. For these reasons, blends containing ASA-2 were examined with further experiments.

The surface gloss of a PC/ASA blend is always lower than that of ASA used in the blend. We examined the surface morphology of a 6/4 PC-1/ASA-2 blend containing 5 phr PMMA as a compatibilizer to understand the reduction of the surface gloss of the blend. As shown in Figure 7, the surface contained a small amount of PC. Because the melt viscosity of PC was higher than that of ASA at the same temperature, ASA was directed to the outer surface under a high shear field, whereas PC with a high viscosity was directed to the inside. For this reason, even though the blend contained 60 wt % PC, a small amount of PC existed at the surface of the blend. PC existing at the surface reduced the surface gloss of the blend. To remove PC existing at the surface of the blend, we controlled the viscosity ratio of PC to ASA by substituting PC-2 for PC-1. Figure 8 shows the surface mor-

phology of 6/4 PC-2/ASA-2 blends containing 5 phr compatibilizer. Even though PC existing at the surface was reduced as the molecular weight of PC increased, PC still existed at the surface when the blend contained the TMPC-*b*-PMMA copolymer as a compatibilizer. However, PC was not observed at the surfaces of the blends containing PMMA as a compatibilizer. Because TMPC-*b*-PMMA was a better compatibilizer, the dimensional stability of the dispersed phase in the blend containing TMPC-*b*-PMMA might have been better than that of the blend containing PMMA as a compatibilizer. The more effective the compatibilizer



(a)



(b)

Figure 7 SEM microphotographs of 6/4 PC-1/ASA-2 blends: (a) without NaOH etching and (b) with the PC-rich phase etched out with a 30 wt % NaOH aqueous solution. The blends contained 5 phr PMMA as a compatibilizer



(a)



(b)

Figure 8 SEM Microphotographs of 6/4 PC-2/ASA-2 blends: (a) with 5 phr TMPC-*b*-PMMA as a compatibilizer and (b) with 5 phr PMMA as a compatibilizer. The blends were treated with a 30 wt % NaOH aqueous solution to etch out the PC-rich phase existing on the surface

was, the more stable the dispersed phase was. Because the phase separation of the blend containing TMPC-*b*-PMMA was retarded during injection molding, PC still remained at the surface of the blend. When PC was completely removed from the surface of the blend with PC-2 and PMMA as the compatibilizer, the surface gloss of the blend reached a maximum value that was the same as that of pure ASA, as shown in Table II.

CONCLUSIONS

A blend of PC and ASA with high surface gloss and improved interfacial properties was developed for use

in automobile parts. To enhance the interfacial properties between PC and the SAN polymer that formed the matrix in the PC/ASA blend, we used two different types of compatibilizers: TMPC-*b*-PMMA copolymer and PMMA homopolymer. The TMPC-*b*-PMMA diblock copolymer was more effective than PMMA in improving the interfacial properties of the PC/ASA blend. The surface gloss of the PC/ASA blend was always lower than that of the pure ASA included in the blend because of PC existing at the surface of the blend. The molecular weight of PC and the compatibilizer type were controlled to develop a PC/ASA blend with optimum surface gloss. PC existing at the surface of blend was completely removed with high-molecular-weight PC (PC-2) and with PMMA as the compatibilizer. This blend exhibited maximum surface gloss that was the same as that of the pure ASA included in the blend.

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