

Weld-Line Characteristics of Polycarbonate/Acrylonitrile–Butadiene–Styrene Blends. I. Effect of the Processing Temperature

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Received 8 December 2003; accepted 13 July 2004

DOI 10.1002/app.21218

Published online in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: The effects of the processing temperature on the morphology and mechanical properties at the weld line of 60/40 (w/w) polycarbonate (PC)/acrylonitrile–butadiene–styrene (ABS) copolymer blends were investigated. The influences of the incorporation of poly(methyl methacrylate) (PMMA) as a compatibilizer and an increase in the viscosity of the dispersed ABS domain phase were also studied. The ABS domain was well dispersed in the region below the V notch, and a coarse morphology in the core region was observed. When tensile stress was applied perpendicularly to the weld line, the fracture propagated along the weak region behind the weld part; there, the domain phase coalescence was significant because of the poor com-

patibility between PC and styrene–acrylonitrile (SAN). Phase coalescence became severe, and so the mechanical strength of the welded specimen decreased with an increasing injection-molding temperature. The domain morphology became stable and the mechanical strength increased as the viscosity of the domain phase increased or some SAN was replaced with PMMA. That the morphology was well distributed behind the weld line and the mechanical properties of PC/ABS/PMMA blends were improved was attributed to the compatibilizing effect of PMMA. © 2004 Wiley Periodicals, Inc. *J Appl Polym Sci* 95: 689–699, 2005

Key words: compatibilization; morphology; polycarbonates

INTRODUCTION

Polycarbonate (PC) is an engineering thermoplastic material with several important characteristic properties: high toughness, high continuous working temperature, high modulus, and transparency. However, major drawbacks of PC include high melt viscosity and notch sensitivity. The disadvantages of PC can be overcome through blending with various thermoplastics or thermoplastic elastomers; of these, acrylonitrile–butadiene–styrene (ABS) copolymer is the most popular.^{1,2}

The relatively favorable interaction between PC and the styrene–acrylonitrile (SAN) copolymer matrix of ABS apparently makes it possible to produce commercially useful materials without any compatibilizer.^{3–10} However, these uncompatibilized PC/ABS blends undergo significantly dispersed phase particle coalescence in the melt state under certain molding conditions, and this leads to a significant deterioration of the properties of the blend.^{11–13} From this point of view, compatibilization is thought to enhance the adhesion between the blend components of PC/ABS

blends.^{14–18} Poly(methyl methacrylate) (PMMA) has been reported as a compatibilizing agent for PC/ABS blends.^{16–18}

The injection molding of large and complex parts requires the use of multigated molds and inserts. The weld line is the visual and structural defect that occurs when two melt flow fronts meet because of either multigated molds (cold weld) or the splitting and rejoining of flow that occurs around inserts (hot weld).

The weakness of the welded region for homopolymer systems arises from the existence of a V notch, molecular orientations induced by the fountain flow, and poor intermolecular diffusion at the weld lines,^{19–29} but it can be minimized by the adjustment of molding conditions such as the melting temperature.

For polymer blends, the problem is much more complex. With the effects of processing conditions, the morphology of the dispersed domain and the interaction parameters between constituent components should be considered. The key is stabilization of the domain morphology at the weld line. The rheological properties of the blend components and the compatibilization are important factors that control the morphology of the dispersed phase at the weld line.^{29–38}

Shieu et al.³² examined the microstructure and tensile strength of the welded zones of PC/ABS blends prepared by two different welding techniques, that is,

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hot-plate welding and vibration welding. The morphology of the weld zone in the hot-plate-welded specimen was highly laminar and oriented, whereas a much more homogeneous microstructure was produced by vibration welding. The welded joint made with the hot-plate technique retained only 30% of the unwelded tensile strength, whereas 80% of the strength was retained in the case of vibration welding. Mekhilef et al.^{36,37} studied the weld-line strength of high-density polyethylene/PC blends. In their study, the degree of bonding near the interface predicted by model based on diffusion and the Flory–Huggins theory of the free energy of mixing was compared with the experimental results. They reported that the weld-line strength of an 80/20 PC/polyethylene blend system increased with an increasing injection-molding temperature between 220 and 300°C. Semba and Hamada³⁹ observed etched samples of PC/ABS injection moldings with scanning electron microscopy. They reported that a weld part with a fine dispersion of ABS was found in the area below the V notch and that the interface between the weld part and the part behind the weld part was the weakest part.

Although it is generally known that the stabilization of the domain morphology would be beneficial to the weld-line strength, there has been little study on the effects of the processing conditions on the morphology and mechanical properties of the welded regions of partially miscible polymer blends. The goal of this study was to determine the effects of the injection-molding temperature on the mechanical properties and morphology at the weld line of 60/40 (w/w) PC/ABS blends. The influences of an increase in the viscosity of the dispersed phase and the incorporation of a compatibilizer into the blends were compared.

EXPERIMENTAL

Materials

ABS was used in the form of a mixture of SAN and ABS graft rubber concentrate (g-ABS). A commercial grade of bisphenol A polycarbonate, two grades of SAN copolymer, g-ABS rubber concentrate, and PMMA were used in this study. PC was in a granular form (Teijin Chemicals, Tokyo, Japan). SAN and a g-ABS rubber concentrate powder were obtained from Cheil Industries, Inc. (Seoul, Korea). PMMA was obtained from LG Chem, Ltd. (Seoul, Korea). The compositions and suppliers of the polymers used in this work are shown in Table I.

Preparation of the polymer blends

PC was dry-blended with g-ABS, SAN, and PMMA to a composition of 60/12/28 w/w/w PC/g-ABS/(SAN–PMMA) before being fed into the hopper of a

TABLE I
Some Characteristics of the Materials

Material	M_w	Characteristics	Supplier
SAN10	95,000	AN content: 24.5 wt %	Cheil Industries
SAN19	192,000	AN content: 27.5 wt %	Cheil Industries
		Core: PBD (58%), shell:	
g-ABS	—	SAN	Cheil Industries
PMMA	103,000	—	LG Chem
PC	25,000	—	Teijin

M_w = weight-average molecular weight; PBD = polybutadiene.

40Φ (length/diameter ratio = 34.5) Berstorff intermeshing, corotating, twin-screw extruder operating at 250 rpm. The temperature ranged from 230°C near the hopper to 250°C at the die exit. The blend was cooled by water at room temperature, pelletized, and stored.

To characterize the properties of the ABS phase in the PC/ABS blend, we made the ABS phase blend as follows. g-ABS was dry-blended with SAN and PMMA to a composition of 12/28 w/w g-ABS/(SAN–PMMA) before being melt-blended in the same extruder. The temperature ranged from 200 to 220°C.

Melt rheology

Capillary rheometry was carried out for the component polymers as well as blends at 240–280°C in a Rosand D10B advanced rheometer system (Malvern, UK) at different shear rates. The Rabinowitsch correction was performed.

Injection molding

The prepared pellets were dried in an air-circulating oven at 80°C for more than 12 h to remove residual moisture. Then, the dried pellets were fed into an injection-molding machine (IDE140ENII, LG Cable, Korea) with a clamping force of 100 tons. The PC/ABS blends were injection-molded at three different injection-molding temperatures (240, 260, and 280°C). A double-gate mold was used to provide a weld line (cold-weld type) in the specimen.

Mechanical properties

The tensile properties of injection-molded specimens were obtained with an Instron 4467 universal tensile machine (Canton, MA) with a crosshead speed of 5 mm/min at room temperature.

The specimen for the impact properties was prepared according to ASTM D 256 (6.4 mm × 3.2 mm × 12.8 mm) with the same injection-molding machine used for the tensile specimen. The impact strength was

measured with a Toyoseiki impact tester (Tokyo, Japan) at room temperature.

Morphological measurements

A small rectangular strip from each sample was placed in liquid nitrogen for more than 10 min and then was fractured manually. The fractured surfaces were subjected to a chemical treatment, by which the ABS component was etched out. The solution for the etching treatment was a mixture of sulfuric acid (240 g), distilled water (60 g), and potassium dichromate (10 g). The treatment time was 10 min at a solution temperature of 75°C. After the etching treatment, the specimens were washed in water. The etched-out surfaces were then treated with a gold-palladium layer and examined with a JEOL JSM-840A scanning electron microscope (Tokyo, Japan).

Samples cut from the mold gate and weld zones of the polymer blends containing rubber particles were trimmed into a shape suitable for ultramicrotomy and then stained with an aqueous OsO₄ solution overnight. Slices less than 80 nm thick were cut with an ultramicrotome from the sample transverse direction to the flow of the polymer blends. Thin sections were further stained with RuO₄ vapor. The microstructure of the weld zone was studied with a JEOL JEM-2000EXII transmission electron microscope.

RESULTS AND DISCUSSION

Melt viscosity of PC and ABS

It is generally accepted that the dispersed domain particles become larger if the viscosity ratio of the blend components increases above unity or decreases below unity. Because the viscosity ratio is also an important factor that controls the morphology of the dispersed phase at the weld line, the melt viscosities of the matrix (PC) and domain (ABS) phase were measured with a capillary rheometer. Figure 1 gives a plot of the melt viscosity versus the shear rate for PC and three kinds of ABS blends [12/28 g-ABS/(SAN10-SAN19)], for which the ratio of the low-molecular-weight SAN copolymer (SAN10) to the high-molecular-weight SAN copolymer (SAN19) was changed, at three temperatures (240, 260, and 280°C). The viscosities of PC at a high shear range at 240 and 260°C were not obtainable because of the pressure-transducer limits of the rheometer. ABS showed significant shear thinning in comparison with PC, and this was indicative of a shear-sensitive material. As the average molecular weight of SAN increased, the viscosity of the ABS domain increased.

The effect of the temperature on the viscosity of PC and ABS was examined. Figure 2 presents a viscosity-temperature plot at a shear rate of 100 s⁻¹. The change

in the viscosity of PC was more sensitive to temperature than that of ABS. A plot of the viscosity ratio versus the temperature at different shear rates is shown in Figure 3 for three kinds of ABS blends containing different amounts of high-molecular-weight SAN19. Because some of the PC viscosities were not obtainable, the viscosity ratios corresponding to the points were also missing. The viscosity ratio decreased with the shear rate at a constant temperature but increased with the melting temperature at a constant shear rate. This tendency was pronounced for low-shear-rate and high-molecular-weight SAN. This can be explained by the distinct shear thinning of ABS at a high shear rate and by the fact that the viscosity of PC decreased more stiffly with increasing temperature than that of ABS. These results agree with earlier observations.⁴⁰ Thus, at this stage, it appears that the mixing should be carried out at a low shear rate and a high temperature to obtain effectively dispersed and distributed domains in PC/ABS blends, as long as we do not consider the fact that the compatibility between PC and SAN decreases at high temperatures. This result is the opposite of later results.

The viscosities of ABS/PMMA blends [12/28 g-ABS/(SAN10-PMMA)] were measured at 240, 260, and 280°C. The blends also showed significant shear thinning, and the viscosity of the ABS/PMMA blends increased as SAN10 was replaced with PMMA. Figure 4 presents viscosity-temperature plots of ABS blends with high-molecular-weight SAN and ABS/PMMA blends at shear rates of 100 and 1000 s⁻¹. The viscosity of the ABS/PMMA blend was lower than that of the ABS blend with high-molecular-weight SAN when the content of PMMA was equal to that of high-molecular-weight SAN. Furthermore, the viscosity of the ABS/PMMA blend was more sensitive to temperature in the investigated range. Therefore, the value of the viscosity ratio (with ABS or ABS/PMMA as the domain and PC as the matrix), of the PC/ABS/PMMA blend was lower than that of the PC/ABS blend with high-molecular-weight SAN when the content of PMMA was equal to that of high-molecular-weight SAN19. On the basis of the previous results concerning the viscosity ratio, it is probable that the dispersion state of the domain phase can be better in a PC/ABS blend system with high-molecular-weight SAN than in a PC/ABS/PMMA blend, if we ignore the compatibility problems of the component polymers.

Weld-line strength and morphology of the PC/ABS blends

Figure 5 shows the influence of the content of high-molecular-weight SAN on the tensile and impact strength at the weld line of PC/ABS blends molded at three different injection-molding temperature. The tensile strength at the weld line increased as the con-

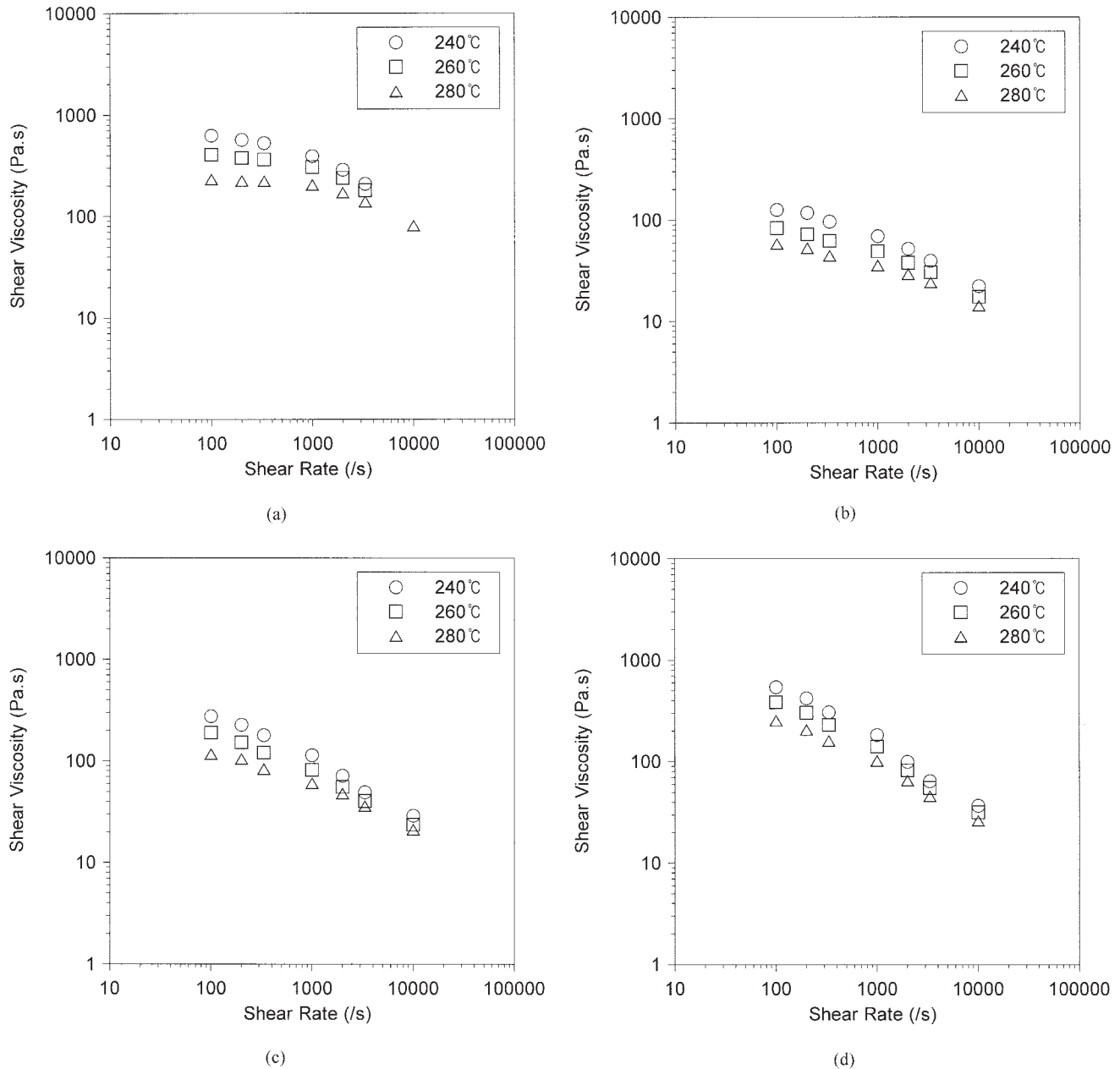


Figure 1 Plots of the viscosity versus the shear rate for PC and ABS [12/28 g-ABS/(SAN10–SAN19)] blends at 240, 260, and 280°C: (a) PC, (b) ABS with 28/0 SAN10/SAN19, (c) ABS with 14/14 SAN10/SAN19, and (d) ABS with 0/28 SAN10/SAN19.

tent of high-molecular-weight SAN increased at all injection-molding temperatures tested. However, the extent of the increase in the weld impact strength with the content of high-molecular-weight SAN depended on the injection-molding temperature. At an injection-molding temperature of 240°C, the weld impact strength increased from 10.0 to 16.7 kg cm/cm as the content of high-molecular-weight SAN increased from 0 to 28%. However, at 280°C, only a slight increase was observed from 10.0 to 11.4 kg cm/cm.

Both the weld tensile strength and weld impact strength decreased as the injection-molding temperature increased from 240 to 280°C. This contrasted with

the study of Mekhilef et al.,^{36,37} in which the weld-line tensile strength of an 80/20 PC/polyethylene blend increased as the injection-molding temperature increased between 220 and 300°C. This opposite result can be explained by the morphology at the weld line of the PC/ABS blend.

The fractured cross sections of the weld part of a 60/12/28 PC/g-ABS/SAN10 blend along the flow direction are shown in Figure 6. Ditches were etched out of part of the rubber component. A flat part without ditches just at the weld line and a clear fountain flow behind the weld line could be observed. This was similar to the results of Semba and Hamada.³⁹ In their

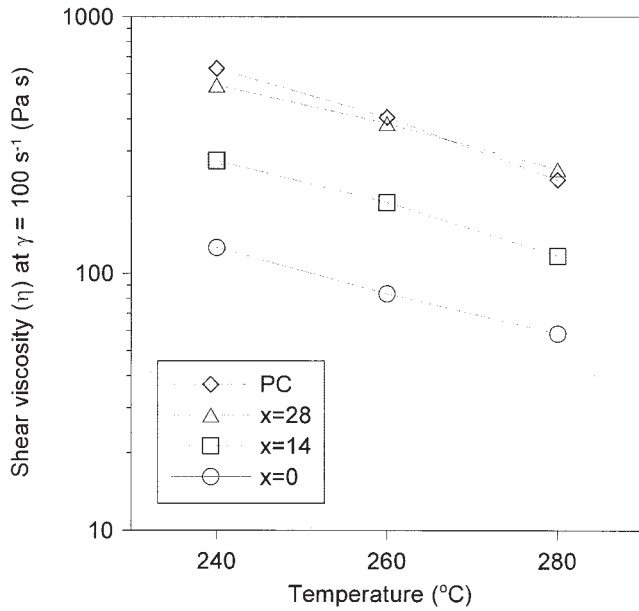


Figure 2 Plots of the shear viscosity versus the temperature for PC and ABS (12/28 - x/x g-ABS/SAN10/SAN19 blends at a shear rate of 100 s^{-1}).

study, they found a very fine dispersion of ABS at the skin layer of the flow front and the weld part below the V notch because of the high extensional stress at the skin layer. They also reported that, because of the short relaxation time of PC and the fine dispersion of PC and ABS, molecular chains of PC became entangled across the weld line in a short time and the weld line disappeared. The interface between the weld part and the part behind the weld part was the weakest part.

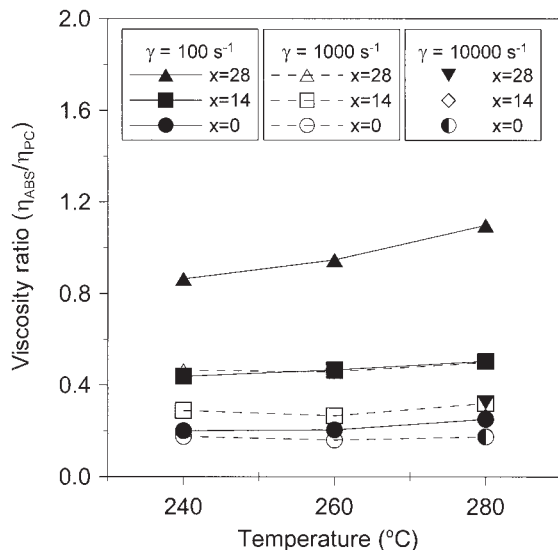


Figure 3 Plots of the viscosity ratio ($\eta_{\text{ABS}}/\eta_{\text{PC}}$) versus the temperature at shear rates of 100, 1000, and $10,000 \text{ s}^{-1}$ for 12/28 - x/x g-ABS/SAN10/SAN19 blends.

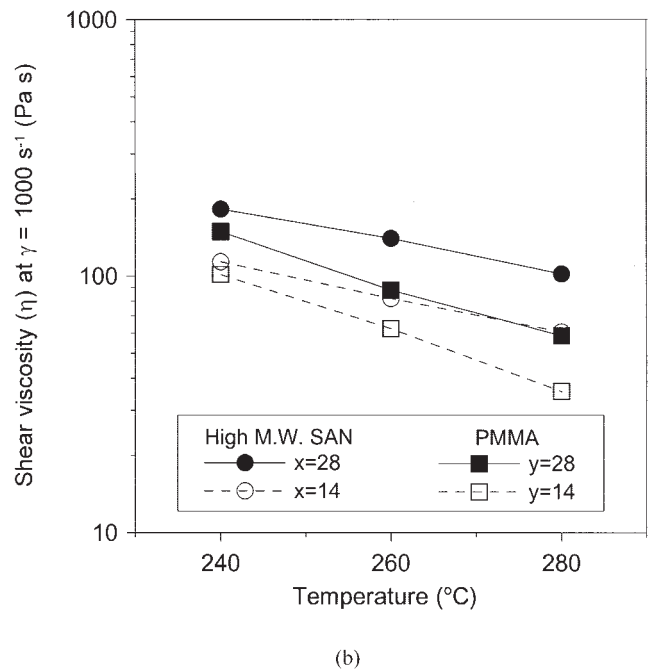
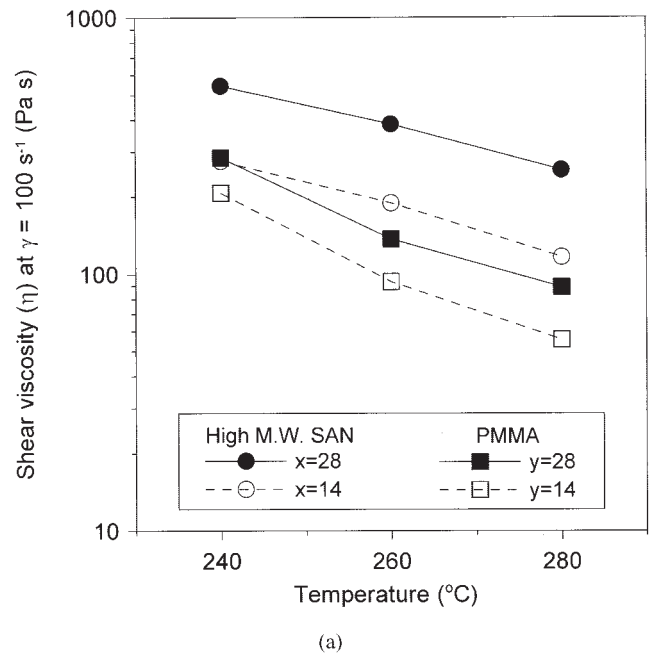


Figure 4 Plots of the shear viscosity versus the temperature for 12/28 - x/x g-ABS/SAN10/SAN19 and 12/28 - y/y g-ABS/SAN10/PMMA blends at shear rates of (a) 100 and (b) 1000 s^{-1} .

When the temperature was increased, molecular chains could easily diffuse and entangle across the weld line. However, severe domain phase coalescence at the core region was expected because of the poor miscibility between the PC and SAN copolymer at high temperatures.¹¹⁻¹³ Therefore, the difference in the morphology between the weld line and core region was expected to be distinct.

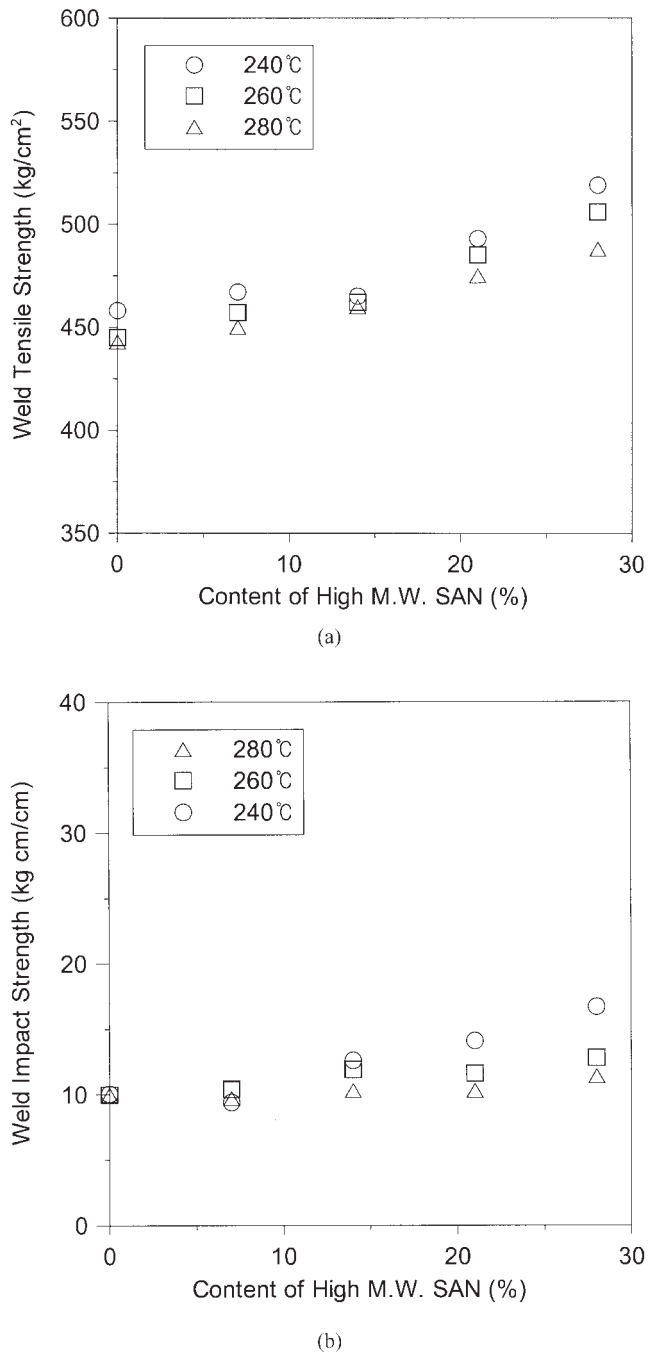


Figure 5 Plots of the weld-line strengths of PC/ABS blends molded at injection-molding temperatures of 240, 260, and 280°C versus the content of high-molecular-weight SAN copolymer: (a) weld tensile strength and (b) weld impact strength.

Figure 7 shows the cross section of the weld part of a 60/12/28 PC/g-ABS/SAN10 blend along the flow direction after tensile testing. The etching method was also applied to this cross section. The fracture propagated along the part behind the weld line. It is not clear whether the fracture occurred at the interface between the weld part and the other part, as reported

by Semba and Hamada.³⁹ However, the crack clearly did not propagate along the weld line.

Figure 8 shows TEM micrographs of 60/12/28 PC/g-ABS/SAN blends at the gate part and the fractured

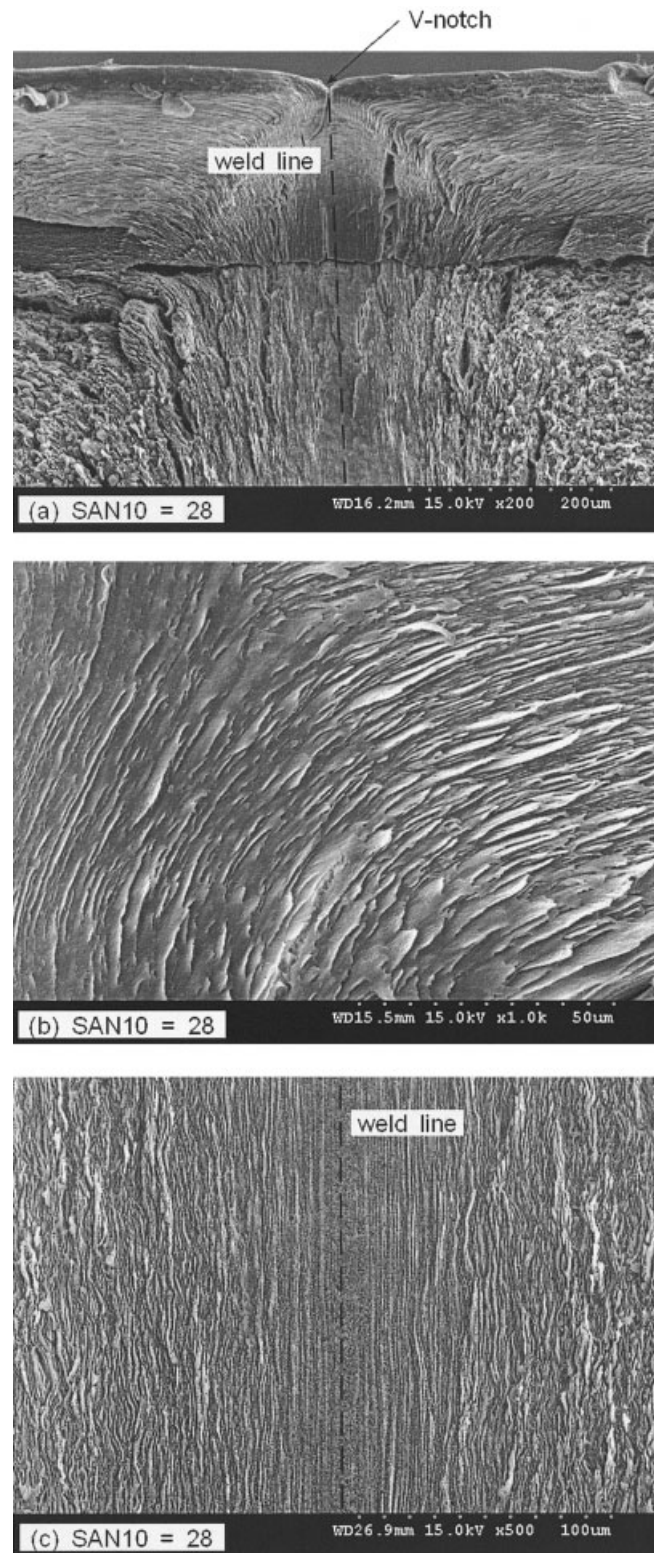


Figure 6 (a) SEM photograph of the cross section at the weld part of a 60/12/28 PC/g-ABS/SAN10 blend molded at 240°C before tensile testing and (b,c) magnifications of the photograph.

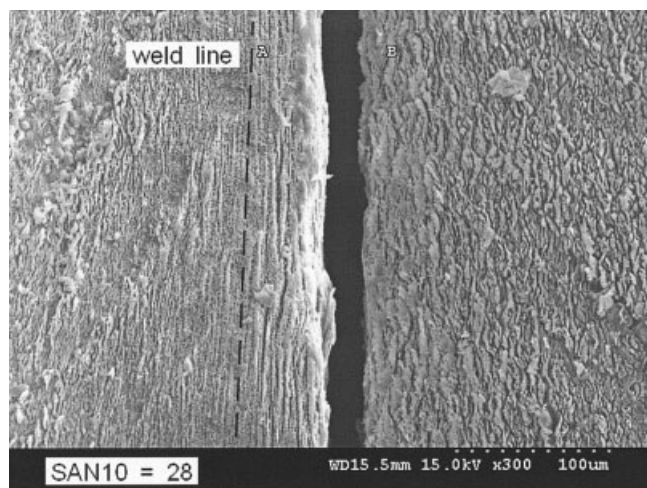


Figure 7 SEM photographs of the cross section at the weld part of a 60/12/28 PC/g-ABS/SAN10 blend molded at 240°C after tensile testing.

weld region molded at the injection-molding temperatures of 240 and 280°C. At the gate part, the PC/ABS blend had a fairly well-distributed morphology, regardless of the molecular weight of SAN and the injection-molding temperature because of the high shear rate during injection molding and rapid cooling rate after injection at the gate part [see Fig. 8 (a–c)]. The morphology at the skin layer of the flow front or just at the weld line should have been similar to that at the gate part. However, severe phase coalescence of the ABS domain was observed at the fractured region near the weld line of the PC/ABS blend with low-molecular-weight SAN10 molded at 240°C [see Fig. 8(d)]. In the case of ABS with high-molecular-weight SAN19, the dispersion state of the ABS domain behind the weld part was enhanced [see Fig. 8(e)]. This could be the result of an increased domain-to-matrix viscosity ratio due to the increase in the content of high-molecular-weight SAN. However, considerable domain phase coalescence still existed. This indicated that the increase in the viscosity of the ABS domain was not enough to stabilize the morphology near the weld line. Phase coalescence became more significant as the injection-molding temperature increased to 280°C [see Fig. 8(f)]. This contrasted with the expectation that a finer domain could be formed with an increase in the processing temperature because of the increased domain-to-matrix viscosity ratio. This severe coalescence might be due to the decreased compatibility between PC and SAN at high temperatures.^{11–13} This was the weak part at which the crack, initiated from the V notch at the surface, propagated along.

These results suggest that the compatibility between the component polymers, rather than the viscosity ratio, plays an important role in the domain morphol-

ogy of PC/ABS blends near the weld line. The mechanical properties of the specimen with a weld line was determined not by the morphology at the weld line but by the morphology at the part behind the weld line. The change in the morphology near the weld line was consistent with the results of the mechanical properties at the weld line. As previously described, PC and ABS were mixed strongly at the skin layer and the weld line even at high temperatures. However, severe domain phase coalescence existed in the core region. Thus, there was a large difference in the morphology between the weld line and the region behind the weld. It is probable that a crack initiated from the V notch at the surface could propagate along the weaker part behind the weld line. Then, the welded specimens molded at high temperatures could have low mechanical strength.

The cross sections of the weld part of a 60/12/14/14 PC/g-ABS/SAN10/PMMA blend along the flow direction before and after tensile testing are shown in Figure 9. The fracture propagated along the part behind the weld line. TEM micrographs near the weld line of PC/ABS blends compatibilized with PMMA are shown in Figure 10. Because of the difficulty encountered in distinguishing the PMMA phase via transmission electron microscopy, proof of the location of PMMA at the interface was not presentable. It was observed that PMMA resided at the interface of PC and SAN containing 31% AN. However, it was completely miscible with SAN containing 23% AN.¹⁶ Furthermore, it is generally known that SAN/PMMA blends are miscible when the weight fraction of AN monomeric units in the SAN copolymer is between 9 and 30%.^{41–43} In this study, with the SAN10 copolymer containing 24.5% AN, PMMA could be incorporated into the ABS phase. TEM micrographs of the specimens molded at the injection-molding temperature of 240°C showed a fine dispersion of the ABS–PMMA domain in the PC matrix when SAN10 was partially replaced with PMMA [see Figs. 8(d) and 10(a,b)]. As the injection-molding temperature increased to 280°C, significant phase coalescence of the ABS–PMMA domain was also observed [see Fig. 10(c)]. However, the degree of phase coalescence was less severe than that in the PC/ABS blend with low-molecular-weight SAN10 molded at 240°C [see Fig. 8(d)] or in the blend with high molecular SAN19 molded at 280°C [see Fig. 8(f)]. This contrasted with the result that the viscosity of the ABS/PMMA blend was lower than that of ABS with high-molecular-weight SAN, and so the dispersion state of the domain phase could be better in the ABS system with high-molecular-weight SAN. This opposite result occurred because we ignored the changes in the compatibility between the component polymers. Therefore, we can conclude that PMMA plays an important role in compatibility enhancement in PC/ABS blends. The influence of an

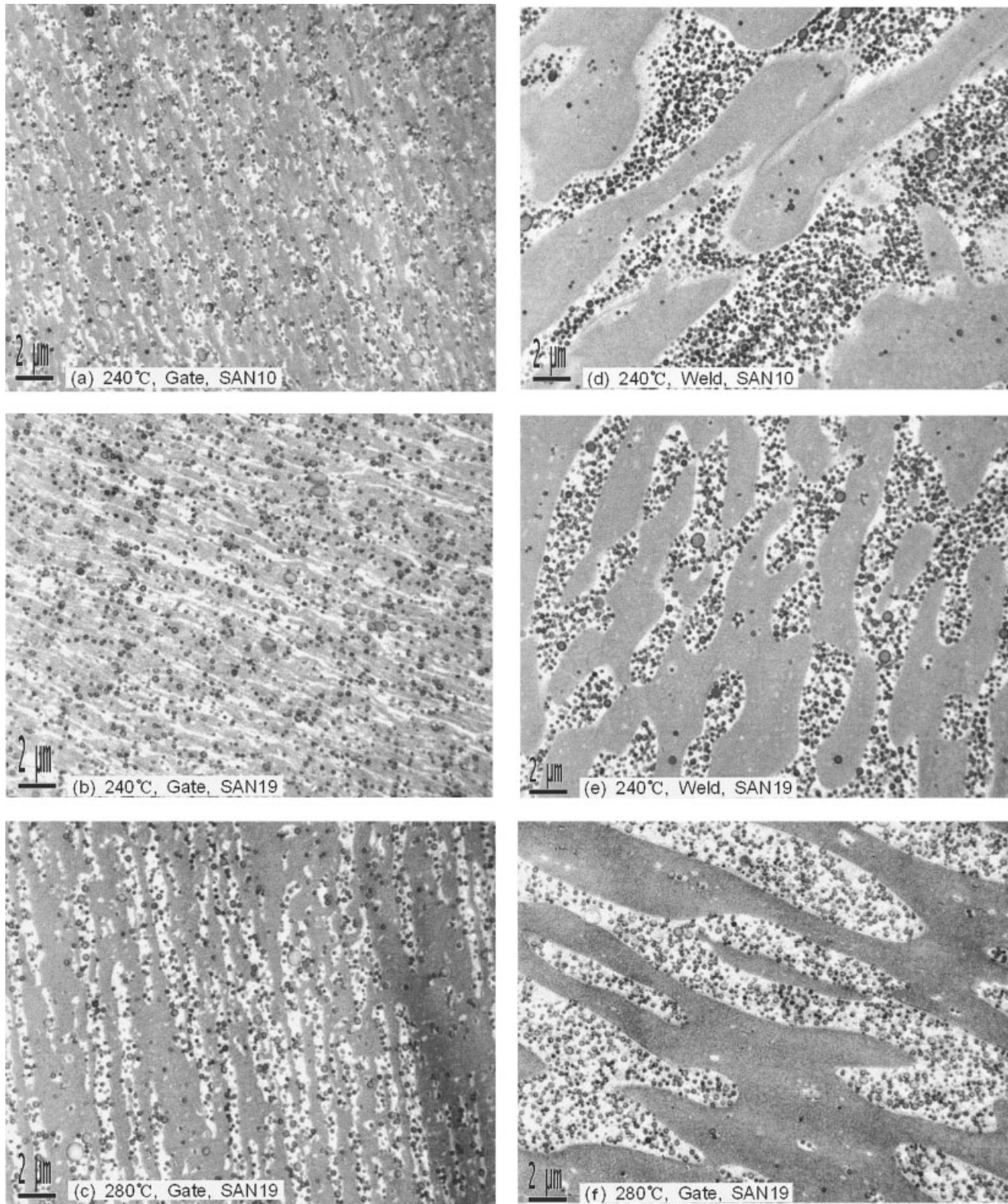


Figure 8 TEM images of 60/12/28 PC/g-ABS/SAN blends at the gate part and weld region molded at injection-molding temperatures of 240 and 280°C: (a) 240°C, gate, and SAN10; (b) 240°C, gate, and SAN19; (c) 280°C, gate, and SAN19; (d) 240°C, weld, and SAN10; (e) 240°C, weld, and SAN19; and (f) 280°C, weld, and SAN19.

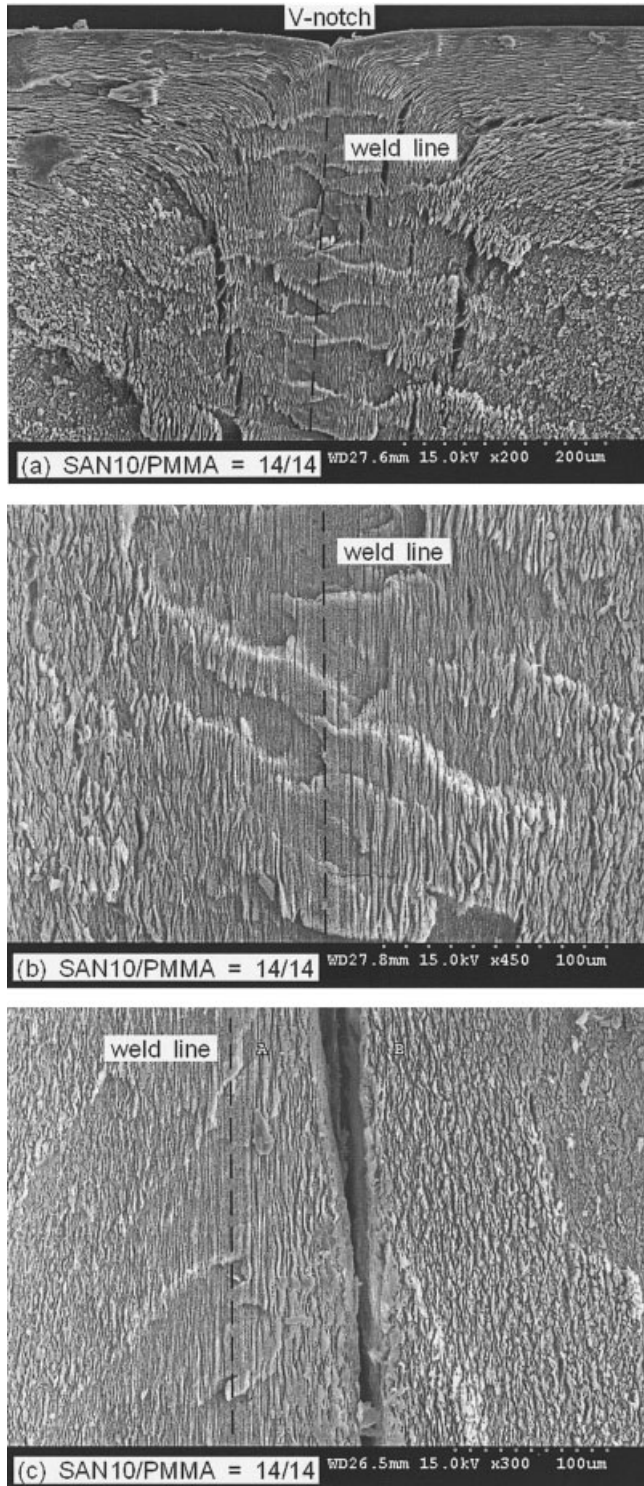


Figure 9 SEM photographs of the cross section at the weld part of a 60/12/14/14 PC/g-ABS/SAN10/PMMA blend molded at 280°C along the flow direction: (a,b) before tensile testing and (c) after tensile testing.

increase in the injection-molding temperature on the morphology of PC/ABS/PMMA blends could be interpreted as the result of partial miscibility and lower critical solution temperature behavior of PC/PMMA and PMMA/SAN blends.

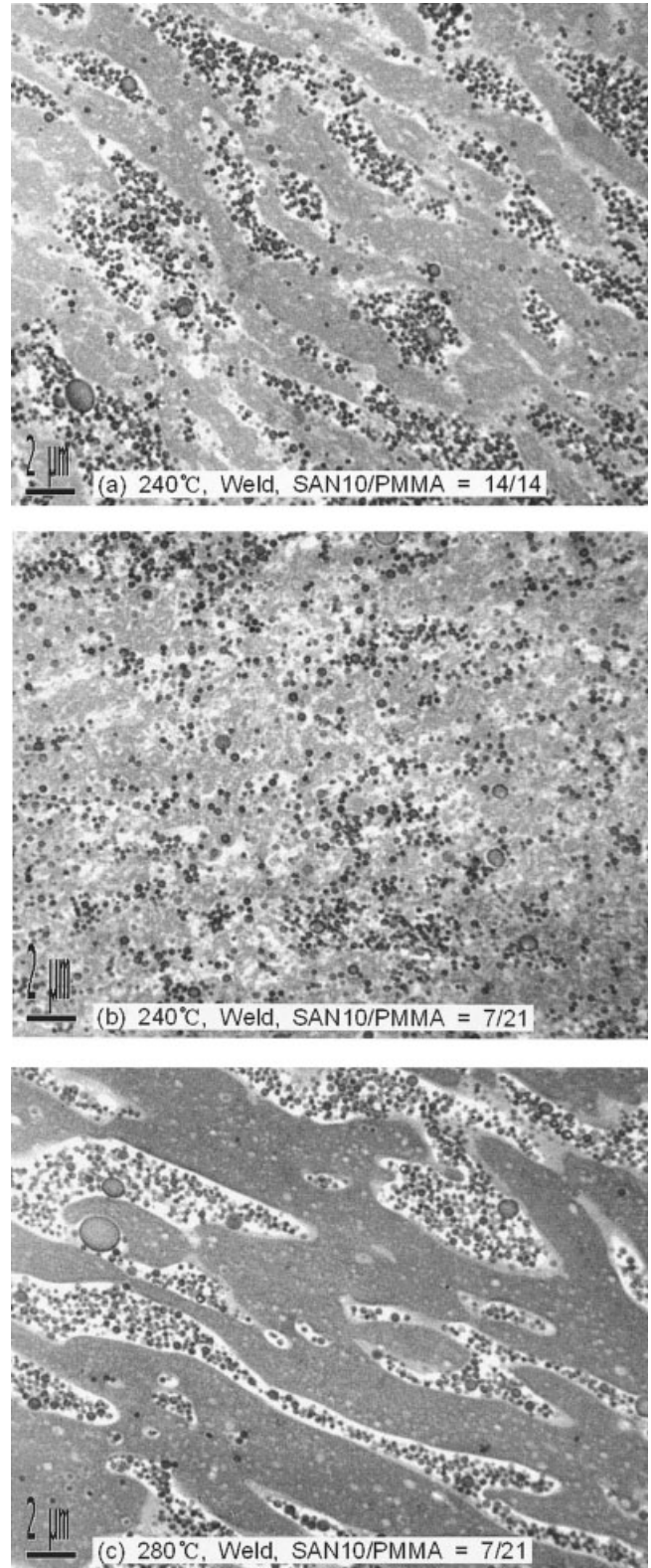


Figure 10 TEM images of the weld regions of 60/12/28-y/y PC/g-ABS/SAN10/PMMA blends molded at injection-molding temperatures of (a) 240°C ($y = 14$), (b) 240°C ($y = 21$), and (c) 280°C ($y = 21$).

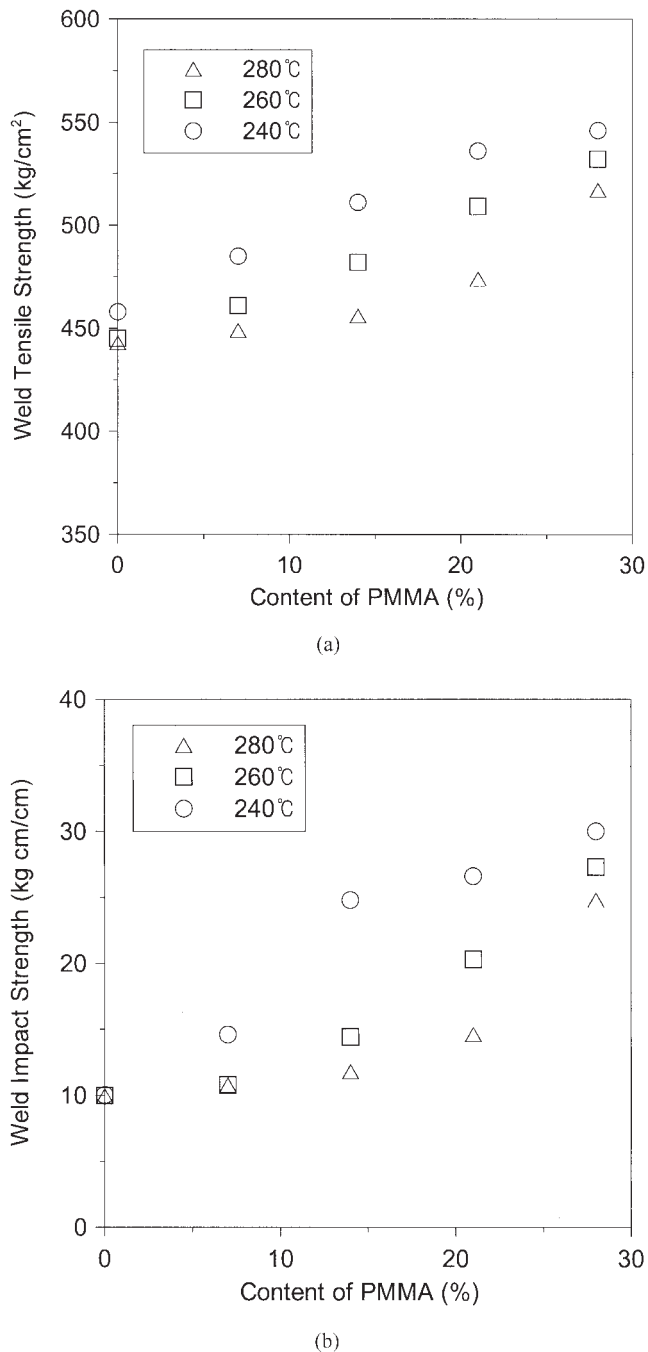


Figure 11 Plots of the weld-line strengths of PC/ABS/PMMA blends molded at injection-molding temperatures of 240, 260, and 280°C versus the content of PMMA: (a) weld tensile strength and (b) weld impact strength.

Figure 11 shows the influence of the PMMA content on the weld-line strength of 60/12/28 PC/g-ABS/(SAN10-PMMA) blends molded at three different injection-molding temperature. As SAN10 was replaced with PMMA, the weld tensile strength increased from 458 to 546 kg/cm², and the weld impact strength increased from 10.0 to 30.0 kg cm/cm, during molding at 240°C. These increases in the weld-line strength of

PC/ABS blends compatibilized with PMMA were much higher than those with high-molecular-weight SAN. The weld-line factor (the strength ratio of the welded and nonwelded specimens) of PMMA is known to be higher than that of SAN.⁴⁴ However, the increase in the weld-line strength of the PC/ABS blend system with PMMA could not be fully attributed to the higher weld-line factor of PMMA. It was, to a great extent, due to the compatibilization effect of PMMA. Because the ABS domain was well dispersed at the weld line and cracks did not propagate along the weld line of PC/ABS blends, we can say that the adhesion state of two flow fronts at the weld line was already enough even though PMMA had not been added. The finer the dispersion state was of the domain at the part behind the weld line, the higher the mechanical strength was.

Similarly to the cases of PC/ABS blends with high-molecular-weight SAN, the weld-line strength decreased as the injection-molding temperature increased from 240 to 280°C. The decrease in the weld-line strength with the injection-molding temperature could also be predicted by the unstable morphology at the weld line (see Fig. 10).

Generally, the injection molding of large and complex parts requires the use of materials with good processability or low melt viscosity, and an elevated processing temperature is needed to injection-mold viscous polymers. However, an increase in the injection-molding temperature results in an unstable morphology near the weld line and decreased mechanical strength of the welded specimen. Thus, to increase weld-line strength, the use of a compatibilizer that results in a finer morphology independent of the processing temperature is definitely needed.

CONCLUSIONS

In this study, we investigated the effects of compatibilizers and processing temperatures on the morphology and mechanical properties at the weld line of PC/ABS blends. An enhancement in the dispersion state of the ABS domain and increased mechanical properties of specimens with a weld line were observed when PMMA was incorporated as a compatibilizer or the viscosity of the ABS domain was increased. The increase in the weld-line strength was more pronounced in blends compatibilized with PMMA than in blends with high-molecular-weight SAN. ABS domain phase coalescence at the part behind the weld line became severe, and the weld-line strength decreased, as the injection-molding temperature increased. The change in the weld-line strength was attributed to the variation of the morphology in the part behind the weld line, along which the fracture propagated.

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