

Effect of compatibilization and ABS type on properties of PBT/ABS blends

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Abstract

Toughened blends of poly(butylene terephthalate) (PBT) with appropriate ABS materials can be prepared without a compatibilizer within limited melt processing situations. As illustrated by transmission electron microscopy, coarsening of uncompatibilized blends occurs under certain molding conditions resulting in a deleterious effect on blend properties. Methyl methacrylate, glycidyl methacrylate (GMA), ethyl acrylate (MGE) terpolymers were shown to be effective reactive compatibilizers for PBT/ABS blends that broaden the processing window and provide improved low temperature impact properties, ABS dispersion, and morphological stability. A twin screw extruder is more effective than the single screw extruder used here for processing these reactive blends. Several ABS types with different rubber contents were examined; generally, materials with very high rubber contents were found to be more beneficial for toughening PBT. Among these high rubber content materials, the ABS material having the lowest melt viscosity was found to be superior for optimizing morphology and impact properties. At least 30% of this material, containing a minimum of 36% rubber, is required for producing toughened blends. Moderate amounts of GMA functionality in the compatibilizer (>5%) and small amounts of compatibilizer in the blend (<5%) significantly improve low temperature impact properties and ABS dispersion. Higher amounts of GMA in the blend increase the room temperature impact strength with little effect on the ductile–brittle transition temperature and increase blend viscosity. © 1999 Elsevier Science Ltd. All rights reserved.

Keywords: Methyl methacrylate-glycidyl methacrylate; Poly(butylene terephthalate); Styrene-acrylonitrile

1. Introduction

Poly(butylene terephthalate) (PBT) is an important engineering thermoplastic with many useful properties; however, neat PBT materials fail in a brittle manner under certain loading conditions such as in notched Izod impact testing [1–31]. The impact properties of PBT can be improved by simple melt blending with appropriate acrylonitrile-butadiene-styrene (ABS) materials [1]. These toughened materials, however, can only be produced within a limited processing range and have an unstable phase morphology; the ABS domains can coalesce during certain low shear conditions (particularly at high temperature when the PBT melt viscosity is low) in the melt resulting in a reduction in mechanical properties.

An earlier article showed that terpolymers of methyl methacrylate (MMA), glycidyl methacrylate (GMA), and ethyl acrylate (EA) (denoted as MGE) are effective

compatibilizers for blends of PBT with styrene/acrylonitrile copolymers (SAN) or ABS materials resulting in an improved SAN or ABS dispersion and morphological stability [2]. Model compound studies were used to show that the carboxyl endgroups of PBT react with the epoxide groups of the GMA repeat units during melt processing to form a graft copolymer at the PBT/ABS interface. This earlier article also examined the effect of MGE composition and content on the generation and stabilization of the morphology of PBT/SAN blends in some detail [2]. Previous work has shown that crosslinking reactions occur in the PBT/ABS/MGE system in addition to the desired graft copolymer formation. It appears that residual acid components in certain emulsion-made ABS materials catalyze ring opening polymerization of the epoxide groups in MGE or other possible mechanisms. In any case, the formation of a gel fraction leads to a reduction in the room temperature impact strength [3]. The purpose of the current article is to explore the effect of MGE composition and content, processing conditions, ABS type, and rubber content on the morphology and mechanical properties of PBT/ABS blends.

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Table 1
Materials used in this study

Designation used here	Supplier designation	Molecular and structural characteristics					Brabender Torque ^a (N m)	Izod impact Strength ^b (J/m)	Source
		Rubber content (wt%)	SAN (free) mol. weight (g/mole)	AN content (wt%)	Graft ratio	Rubber particle size (μm)			
ABS-38 ^c	Lustran	38	$\bar{M}_n = 59\ 000$ $\bar{M}_w = 130\ 000$	30	0.57	0.18	13.4	540	Monsanto
ABS-45-a	Starex	45	$\bar{M}_n = 35\ 000$ $\bar{M}_w = 90\ 000$	25	0.45	0.30	16.5	550	Cheil Industries
ABS-45-b	Starex	45	$\bar{M}_n = 37\ 000$ $\bar{M}_w = 85\ 000$	25	0.60	0.30	22.2	525	Cheil Industries
ABS-45-c	Starex	45	$\bar{M}_n = 35\ 000$ $\bar{M}_w = 72\ 000$	24	0.72	0.12, 0.30 ^d	20.8	460	Cheil Industries
ABS-50	BL-65	50	$\bar{M}_n = 44\ 000$ $\bar{M}_w = 167\ 000$	24	0.40	0.05, 0.20 ^d	23.5	520	Sumitomo-Naugatuck
SAN	Tyrl 25	0	$\bar{M}_n = 77\ 000$ $\bar{M}_w = 152\ 000$	25			6.5	30	Dow Chemical
PBT	Vialox 315								
PMMA	Plexiglas V811 (100)		$\bar{M}_n = 63\ 000$ $\bar{M}_w = 158\ 000$				5.8	50	General Electric
							6.7	–	Rohm&Ilaas

^a Values taken at 250°C and 50 rpm, after 10 min.

^b Room temperature notched Izod impact.

^c All the ABS grades were designated by their weight percent rubber.

^d Bimodal particle size distribution.

Table 2
Characterization of MMA/GMA/EA terpolymers

Designation used here	Composition MMA/GMA/EA (weight ratio of monomer feed)	\bar{M}_n (g/mol)	\bar{M}_w (g/mol)	T_g (°C)
MGE-0 (PMMA) ^a	–	63 000	158 000	108
MGE-1	97/1/2	48 000	85 000	107
MGE-3	95/3/2	40 000	120 000	105
MGE-5	93/5/2	41 000	120 000	104
MGE-10	88/10/2	46 000	190 000	102
MGE-15	83/15/2	41 000	119 000	98
MGE-20	78/20/2	26 000	80 000	92

^a Plexiglas V811 (100) from Rohm and Haas Co.

2. Experimental

Table 1 describes the materials used in this work. The rheological behavior of each material was characterized by measurements of the Brabender torque at 50 rpm and 250°C using a 50 ml mixing bowl. The PBT was supplied by General Electric. Five ABS materials with different characteristics were obtained from various sources and are designated by their rubber content, expressed as weight percent. Three of the materials (ABS-38, ABS-45-a, ABS-50) were used in a previous study of uncompatibilized PBT/ABS blends [1]. These materials are emulsion-made SAN grafted rubber concentrates with small rubber particles in the range of 0.18 to 0.3 μm . ABS-45-c and ABS-50 have broad rubber particle size distributions while the other three contain near monodisperse rubber particles. The amount of SAN chains grafted to the rubber particles for these ABS materials, expressed as a graft ratio, ranges from 0.40 to 0.72 as determined by the manufacturer using procedures described elsewhere [1]. The SAN material used in this study was obtained from Dow Chemical. A commercial poly(methyl methacrylate) (PMMA) product from Rohm and Haas was used as the control, i.e. 0% GMA in the MGE-X compatibilizer series shown in Table 2, where the X represents the weight fraction of GMA in the terpolymer. The MGE materials were designed to have similar

rheological characteristics as that of PMMA; their synthesis and characterization are described elsewhere [2].

Pellets of PBT along with the reactive compatibilizer were cryogenically ground to a powder and dried for 16 h in a vacuum oven at 65°C. The ABS powders were dried for 16 h in a convection oven at 70°C. All components for each blend were thoroughly mixed prior to melt compounding using a single pass extrusion method. Table 3 provides information about the processing conditions and the equipment used to prepare PBT blends with various ABS materials. The single screw extruder used, outfitted with an intensive mixing head, proved to be a very effective compounding device; however, it generally provides less effective mixing than the twin screw extruder used in the case of current blends. The range of processing temperatures is limited by the melting point (220°C) and the degradation processes that occur at temperatures greater than

Table 3
Processing conditions used to prepare PBT/ABS blends

Extrusion conditions			Molding conditions ^a
Extruder	RPM	Temperature (°C)	Melt temperature (°C)
Twin screw ^b	170	220	240
		260	260
		260	240
Single screw ^c	30	230	240

^a Arburg allrounder injection molding machine, mold temperature = 50°C, injection and holding pressure = 50 bar, injection time = 3 sec.

^b Baker-Perkins co-rotating, fully intermeshing twin screw extruder (D = 15 mm), residence time 20 sec.

^c Killion single screw extruder (L/D = 30, D = 2.54 cm) outfitted with a high intensity mixing screw, residence time 2 min.

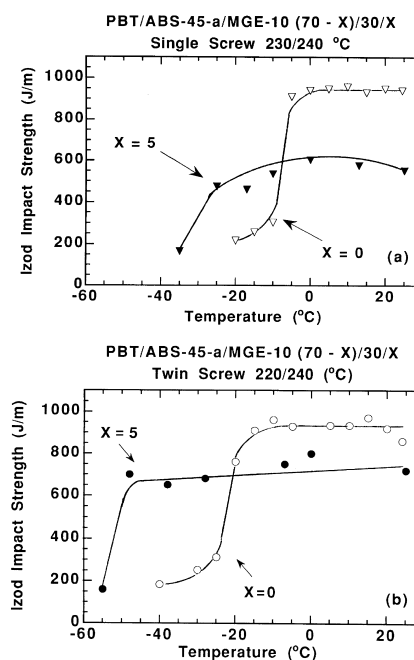


Fig. 1. Effect of temperature on notched Izod impact strength for PBT/ABS-45-a/MGE-10(70-X blends molded at 240°C and extruded at (a) 230°C in a single screw extruder; (b) 220°C in a twin screw extruder.

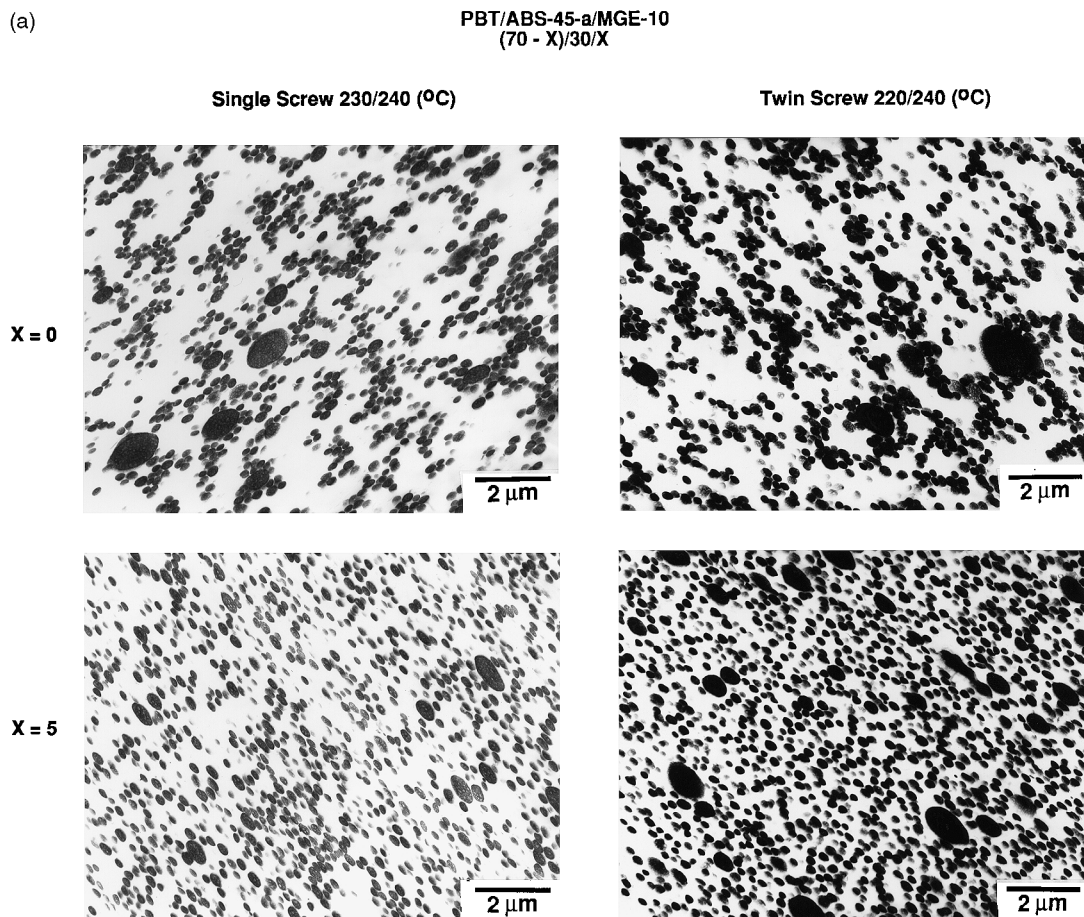


Fig. 2. TEM photomicrographs of PBT/ABS-45-a/MGE-10 (70-X)/30/X blends molded at 240°C and prepared in a single screw extruder at 230°C or a twin screw extruder at 220°C. The rubber of the ABS phase is stained dark by OsO₄ in (a) while the SAN of the ABS phase is stained dark by RuO₄ in (b).

260°C for PBT. The twin screw extruder could be used at a barrel set point of 220°C, as the high shear generates heat that ensures that PBT is fully molten; however, the single screw extruder could not be operated at a set point below 230°C. The temperature profiles for both the extruders and the injection molding machine were kept flat along the barrel. Blends were molded into dog-bone shaped tensile specimens (ASTM D638 type I) and standard Izod bars of 3.13 mm thickness. The mold temperature was kept constant at 50°C.

Notched Izod impact testing was conducted according to ASTM D256 as a function of temperature to determine the ductile–brittle transition temperature. At least five samples, each from the gate- and far-ends of the injection molded bars, were tested at room temperature and in the region of the ductile–brittle transition. At other temperatures fewer samples were tested with the exact number being dictated by the consistency observed. Only gate-end information is reported here as differences between gate- and far-end specimens were typically insignificant. An Instron was used for tensile testing in accordance with ASTM D638 at a crosshead speed of 5.08 cm/min. An extensometer strain gauge with a 5.08 cm gap was used to obtain the modulus and yield stress values.

The morphology of selected blends was examined using a JEOL JEM 200 cx transmission electron microscope (TEM) at an accelerating voltage of 120 kV. Ultra-thin sections (15–20 nm thick) were obtained by cryo-microtoming, from the center of the molded Izod bars perpendicular to the direction of flow during mold filling, using a Riechert-Jung Ultracut E microtome at – 45°C. These sections were exposed to OsO₄ vapor (stains the rubber particles) for 15 h or RuO₄ vapor (stains the SAN matrix) for 20 min. Under these conditions the stained materials appear as dark domains in the TEM images.

3. Results and discussion

One of the objectives of this work is to explore the effect of MGE composition and content on the toughness and morphology of PBT/ABS blends. However, appropriate blending conditions and extruder type must be established first to ensure that optimal blend properties are achieved. In the next sections, processing conditions, ABS type, and rubber content in the blend will be examined. Once the

(b)

PBT/ABS-45-a/MGE-10
(70 - x)/30/x

Single Screw 230/240 (°C)

Twin Screw 220/240 (°C)

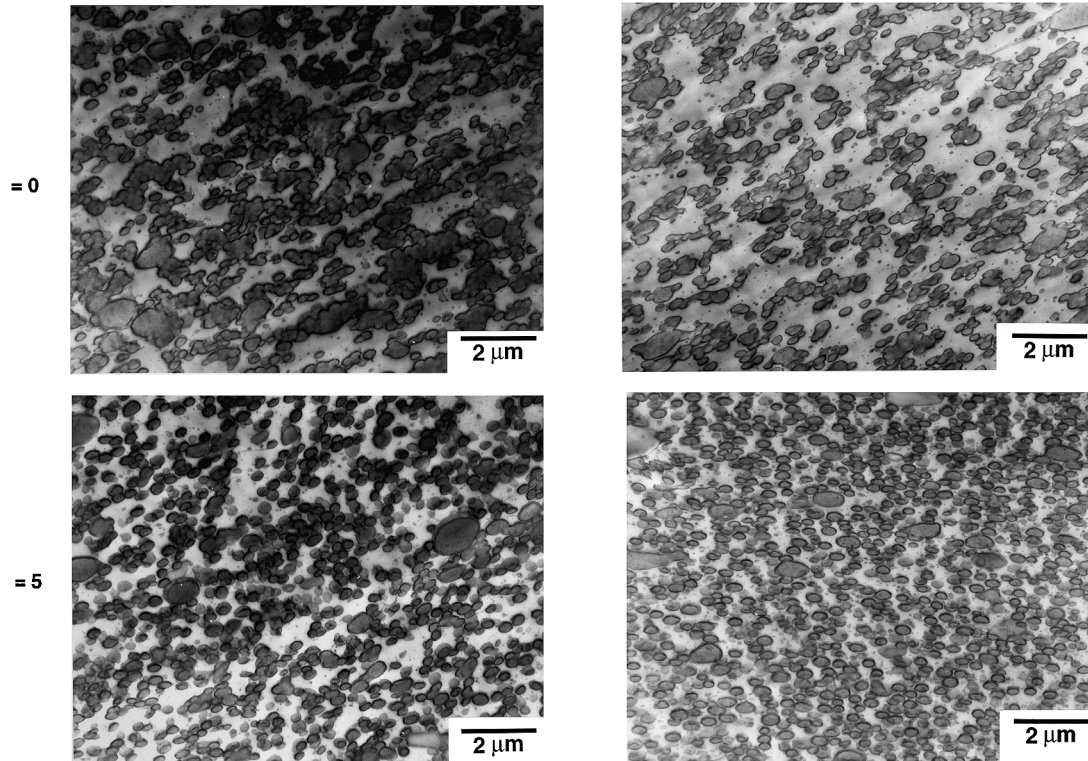


Fig. 2. (continued)

effect of blending protocols are defined, the optimal MGE composition and content will be examined.

3.1. Processing conditions

The notched Izod impact strengths of blends containing 30% ABS with and without the MGE-10 compatibilizer, prepared in the single screw and twin screw extruders, are compared in Fig. 1. Binary blends prepared in the twin screw extruder are tough at lower temperatures than those prepared in the single screw extruder for the processing conditions shown. The addition of 5% MGE-10 reduces the ductile–brittle transition temperature from -8°C to -30°C for blends prepared in the single screw extruder and from -22°C to -53°C for blends prepared in the twin screw extruder. The addition of compatibilizer, however, reduces the room temperature impact strength by 10%–40%. This reduction in impact strength caused by crosslinking reactions involving the MGE epoxide groups, induced by acidic impurities in the emulsion-made ABS, is essentially eliminated when PBT and MGE-10 are blended together prior to adding ABS in a second pass extrusion [3]. Only a single pass extrusion method will be employed in the current work as this minimizes the number of

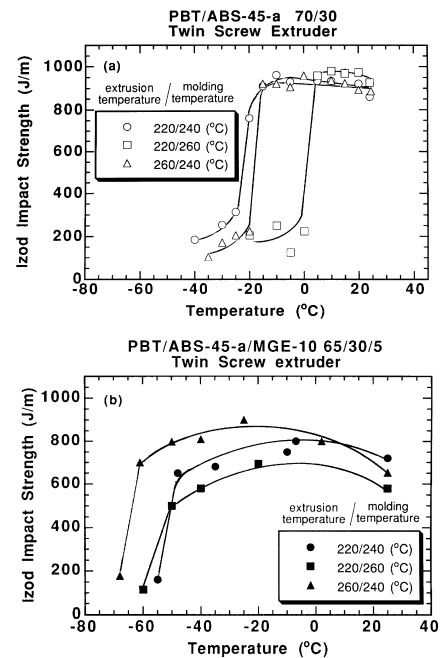


Fig. 3. Effect of temperature on notched Izod impact strength for blends molded and extruded in a twin screw extruder at various temperatures: (a) PBT/ABS-45-a (70/30); (b) PBT/ABS-45-a/MGE-10 (65/30/5).

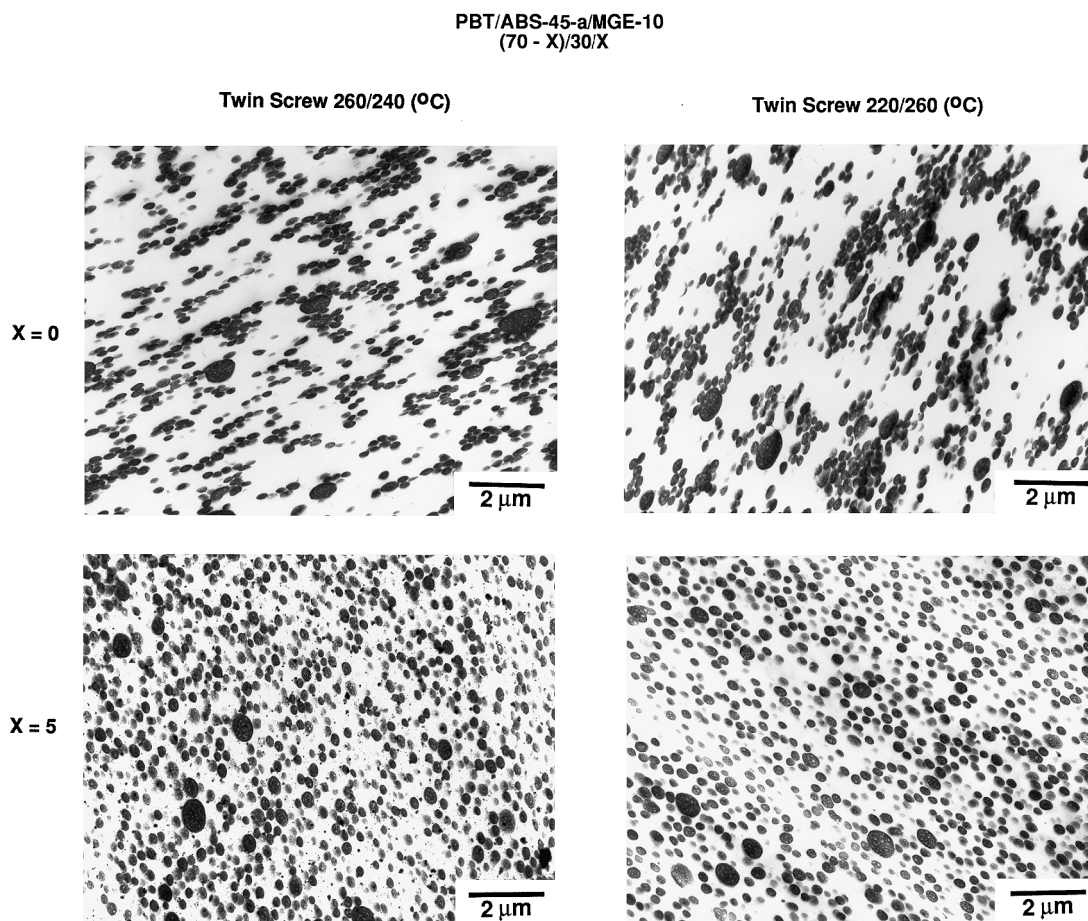


Fig. 4. TEM photomicrographs of PBT/ABS-45-a/MGE-10 (70-X)/30/X blends prepared in a twin screw extruder and molded at various temperatures. The rubber in the ABS phase is stained dark by OsO_4 .

processing steps and is most likely the way such blends would be made in practice.

TEM photomicrographs shown in Fig. 2 provide information about the morphology of these various blends. For the specimens shown in Fig. 2(a), the rubber particles in the ABS appear dark, while in Fig. 2(b) the SAN matrix appears dark as a result of the RuO_4 staining; the latter allows the outline of the entire ABS domains to be seen. The ABS domains appear to be relatively large for uncompatibilized blends; hence, the rubber particles are non-uniformly distributed in the PBT matrix. Addition of 5% MGE-10 to these blends produces a finer dispersion of ABS in the PBT matrix such that only a few rubber particles are located in each domain. There are some rather large ABS domains in the blend prepared in the single screw extruder while the ABS domains are uniformly smaller for the blend made in the twin screw extruder. This is consistent with the more intensive mixing generally found in corotating twin screw extruders compared to single screw extruders.

The toughness of uncompatibilized PBT/ABS blends is rather sensitive to processing conditions as they have a rather unstable morphology. Addition of a compatibilizer like the MGE series described here should, to some degree,

lessen this problem. The effects of extrusion and molding temperatures on the Izod impact strength of PBT/ABS-45-a/MGE-10 blends containing 30% ABS are seen in Fig. 3. When uncompatibilized blends are molded at 240°C, the extrusion temperature does not appear to have a significant effect on the impact properties (Fig. 3(a)). However, these properties deteriorate when the blends are molded at a higher temperature as demonstrated by the increase in ductile–brittle transition temperature from -22°C to 3°C as the temperature of the barrel of the injection molding machine is increased from 240°C to 260°C. This loss of toughness apparently results from coarsening of the ABS phase during molding. Addition of 5% MGE-10 to these blends reduces the ductile–brittle transition temperature while the room temperature impact strength is slightly lowered relative to uncompatibilized blends (Fig. 3(b)). The ductile–brittle transition temperature is lowest (-65°C) when the blends are extruded at 260°C and molded at 240°C. As the MGE-g-PBT graft copolymer provides increased morphological stability, molding at 260°C rather than 240°C is not as deleterious as observed for uncompatibilized blends.

The TEM photomicrographs shown in Fig. 4 reveal little

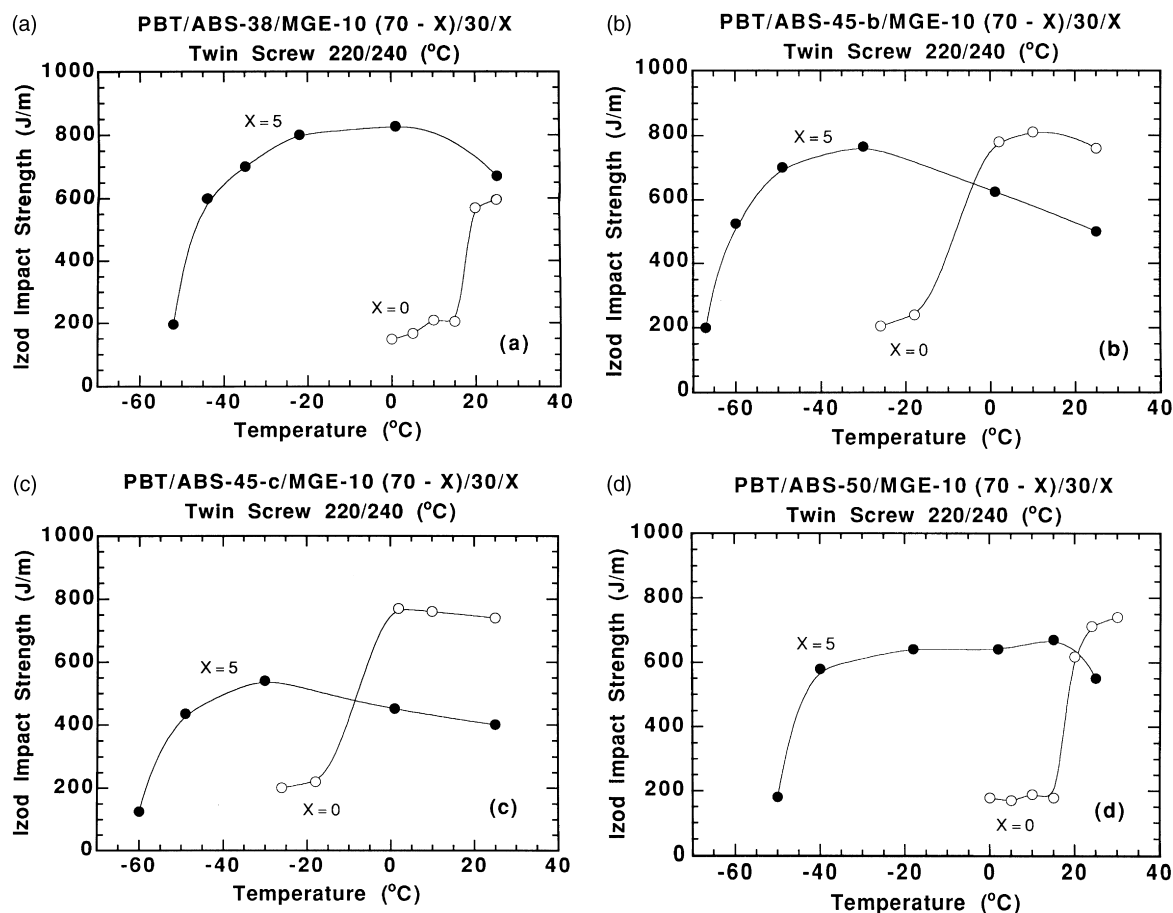


Fig. 5. Effect of temperature on notched Izod impact strength for PBT/ABS/MGE-10 (70-X)/30/X blends prepared in a twin screw extruder at 220°C and molded at 240°C for the following ABS types: (a) ABS-38; (b) ABS-45-b; (c) ABS-45-c; (d) ABS-50.

morphological difference in the blends molded at 240°C when prepared in the twin screw extruder at 260°C or 220°C (Fig. 2). The morphological instability of uncompatibilized blends is particularly apparent during molding at high temperatures where coarsening can occur more rapidly owing to the lower melt viscosity; this effect can be seen by comparing the structure of blends molded at 260°C to those molded at 240°C. The presence of 5% MGE-10 stabilizes the morphology against coalescence during molding at

260°C as suggested by the impact properties shown in Fig. 3(b).

3.2. ABS type

The five emulsion-made ABS materials described in Table 1 (from various commercial sources with different rubber contents, morphological and rheological characteristics) were used to prepare a series of PBT/ABS blends of fixed ABS content (30%) with and without 5% of the MGE-10 compatibilizer. The blends were made in the twin screw extruder at 220°C and were injection molded at 240°C into test specimens. Fig. 1(b) and Fig. 5 show the Izod impact strength as a function of temperature for these blends while Table 4 summarizes their room temperature impact strength and ductile–brittle transition temperatures.

Of the five ABS materials, ABS-38 contains the least amount of rubber. Its binary blend with PBT has a ductile–brittle transition temperature of 18°C. Addition of 5% MGE-10 to this blend reduces the ductile–brittle transition temperature to –47°C and slightly increases the room temperature impact strength.

ABS-45-b is similar to ABS-45-a but has a higher melt viscosity, as indicated by Brabender torque measurements,

Table 4
Impact data for PBT/ABS/MGE-10 (70-X)/30/X blends^a

ABS type	Ductile–brittle transition temperature (°C)		Room temperature Izod impact strength (J/m)	
	X = 0	X = 5	X = 0	X = 5
ABS-38	18	–47	596	670
ABS-45-a	–22	–53	859	720
ABS-45-b	–10	–64	760	500
ABS-45-c	–10	–54	740	400
ABS-50	18	–45	711	550

^a All blends were prepared in the twin screw extruder at 220°C and molded at 240°C.

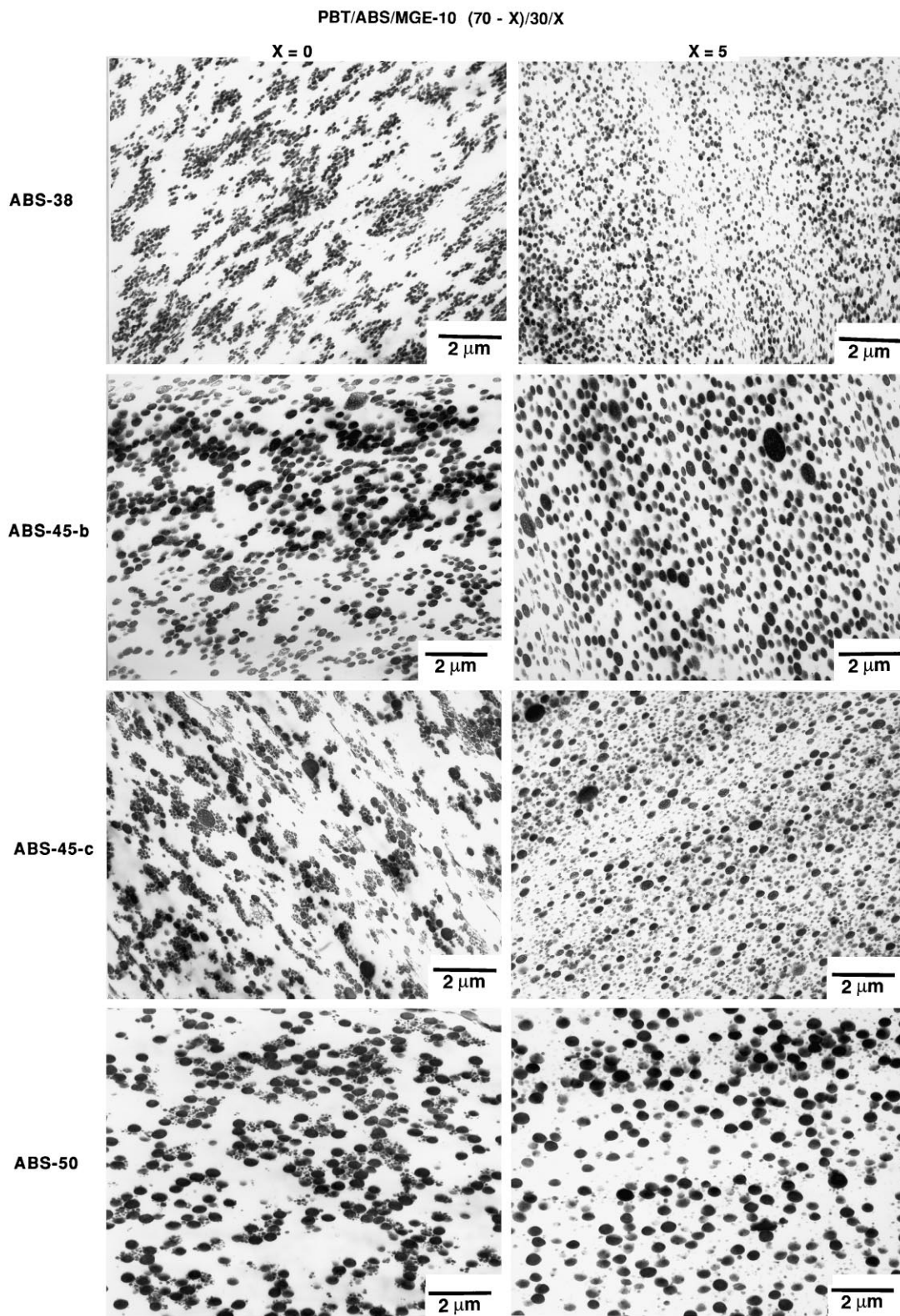


Fig. 6. TEM photomicrographs of PBT/ABS/MGE-10 (70-X)/30/X blends with varying ABS type prepared in a twin screw extruder at 220°C and molded at 240°C. The rubber in the ABS phase is stained dark by OsO₄.

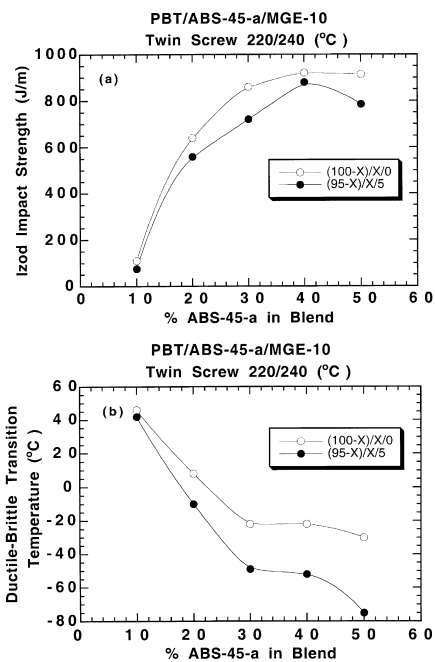


Fig. 7. Effect of ABS content on room temperature notched Izod impact strength (a) and ductile–brittle transition temperature (b) of PBT/ABS-45-a/MGE-10 blends prepared in a twin screw extruder at 220°C and molded at 240°C.

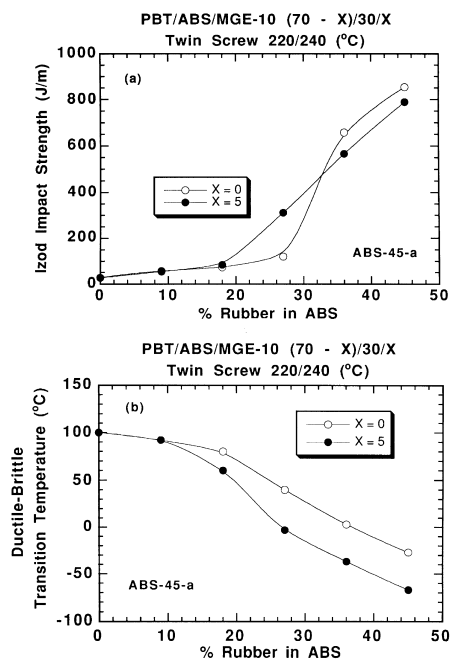


Fig. 9. Effect of ABS rubber content on room temperature notched Izod impact strength (a) and ductile–brittle transition temperature (b) of PBT/ABS-45-a/MGE-10(70-X)/30/X blends prepared in a twin screw extruder at 220°C and molded at 240°C.

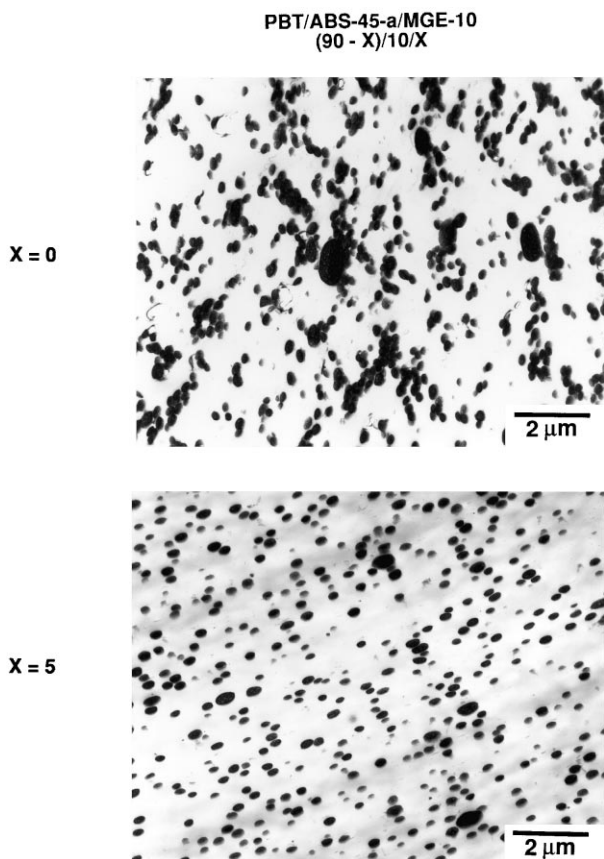


Fig. 8. TEM photomicrographs of PBT/ABS-45-a/MGE-10 (90-X)/10/X blends prepared in a twin screw extruder at 220°C and molded at 240°C. The rubber in the ABS phase is stained dark by OsO₄.

in spite of the higher \overline{M}_w of the free SAN (Table 1) in the latter; evidently the higher graft ratio of the former is responsible for its higher viscosity. As a result of the higher viscosity, the ABS-45-b material is more difficult to disperse into PBT during extrusion compounding; and apparently for this reason, uncompatibilized blends with ABS-45-b have a higher ductile–brittle transition temperature than blends with the lower viscosity ABS-45-a material (Fig. 1(b)). Addition of 5% MGE-10, however, aids in the dispersion of ABS-45-b throughout the PBT matrix resulting in a lower ductile–brittle transition temperature. The presence of compatibilizer also reduces the room temperature impact strength.

Blends based on ABS-45-c have similar impact properties as blends with ABS-45-b (Fig. 5(b)), probably because of the higher viscosity of ABS-45-c or possibly its broad particle size distribution as compared to ABS-45-a. Addition of 5% MGE-10 to this blend lowers the ductile–brittle transition temperature, however, the room temperature impact strength is also reduced significantly.

ABS-50 has the highest rubber content of all the ABS materials used, but its binary blend with PBT has one of the highest ductile–brittle transition temperatures (18°C) in this series of blends (Table 4). ABS-50 has a high melt viscosity, which makes its dispersion into PBT during processing more difficult, and a broad rubber particle size distribution which may affect blend properties. The high rubber content should be beneficial to the impact toughness, however, it was suggested that chemical degradation may be an issue for blends based on ABS-50 [1]. Residual

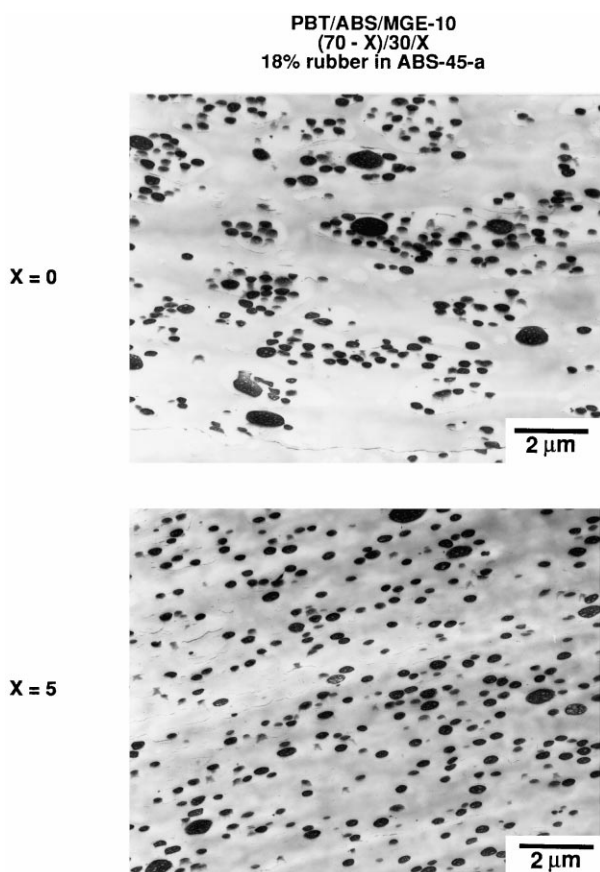


Fig. 10. TEM photomicrographs of PBT/ABS/MGE-10 (70-X)/30/X blends prepared in a twin screw extruder at 220°C and molded at 240°C where the ABS phase (ABS-45-a diluted with SAN) contains 18% rubber. The rubber in the ABS phase is stained dark by OsO₄.

chemicals in ABS-50 were shown to degrade polycarbonate resulting in inferior blend properties [32], there is some evidence that suggests that a similar problem may occur when processing with PBT. Nevertheless, addition of 5% MGE-10 to the blend reduces the ductile–brittle transition

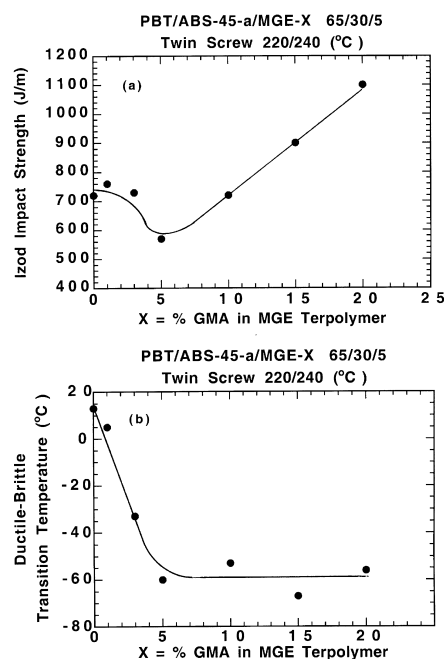


Fig. 11. Effect of GMA content (X) of MGE-X terpolymer on room temperature notched Izod impact strength (a) and ductile–brittle transition temperature (b) of PBT/ABS-45-a/MGE-X (65/30/5) blends prepared in a twin screw extruder at 220°C and molded at 240°C.

temperature to -45°C with a slight reduction in room temperature impact strength.

Fig. 6 shows TEM photomicrographs for all the blends whose properties are listed in Fig. 5. The ABS dispersion within the PBT matrix is relatively poor for all blends without compatibilizer. The materials designated as ABS-45-c and ABS-50 have a bimodal rubber particle size and the smaller population of rubber particles seems to surround the larger rubber particles in the ABS domains of blends based on each of these materials. Addition of 5% MGE-10 to these blends provides a more uniform distribution of rubber throughout the matrix. Even

Table 5
Tensile properties of blends^a

Composition (wt%)	Yield stress (MPa)	Modulus (GPa)	Elongation (%)
PBT	48 ± 0.3	2.4 ± 0.21	165 ± 58
PBT/ABS-45-a/MGE-10			
70/30/0	36 ± 0.1	1.7 ± 0.10	110 ± 59
69/30/1	38 ± 0.1	1.8 ± 0.08	61 ± 32
67/30/3	39 ± 0.3	1.7 ± 0.04	53 ± 30
65/30/5	37 ± 0.2	1.8 ± 0.07	50 ± 6
PBT/ABS-45-a/MGE-X			
65/30/5			
X = 0	39 ± 0.2	1.8 ± 0.08	153 ± 57
X = 1	39 ± 0.2	1.7 ± 0.06	140 ± 53
X = 5	40 ± 0.4	1.8 ± 0.08	114 ± 49
X = 15	39 ± 0.3	1.9 ± 0.05	40 ± 13
X = 20	38 ± 0.4	1.9 ± 0.08	245 ± 7

^a All blends were prepared in the twin screw extruder at 220°C and molded at 240°C. Five specimens of each sample were tested at 5.08 cm/min.

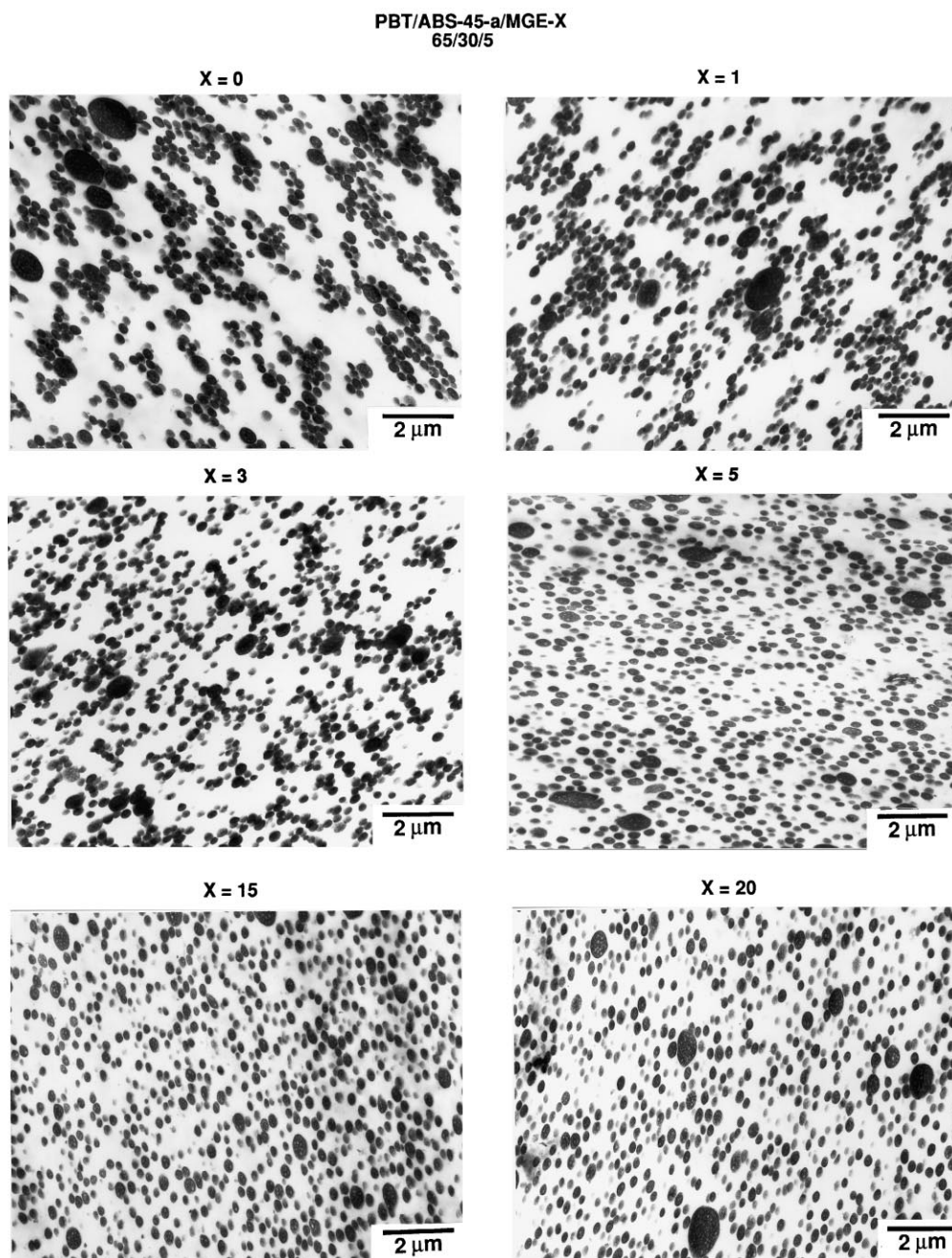


Fig. 12. TEM photomicrographs of PBT/ABS-45-a/MGE-X (65/30/5) blends with a varying amount of GMA in the MGE-X terpolymer prepared in a twin screw extruder at 220°C and molded at 240°C. The rubber in the ABS phase is stained dark by OsO₄.

the smaller rubber particles of ABS-45-c and ABS-50 seem well dispersed.

3.3. Rubber content in the blend

Two series of blends were prepared in order to investigate the effect of rubber content on the impact properties of blends with and without compatibilizer. In the first series, the ABS content of the blend is varied, while in the second the weight fraction of rubber in the ABS is varied keeping the ABS content in the blend fixed at 30%. ABS-45-a was

used in the second series because it has a relatively low melt viscosity and imparts superior impact properties to PBT blends compared to the other ABS materials (Table 4). The Izod impact strength of PBT/ABS-45-a/MGE-10 blends prepared in the twin screw extruder is shown in Fig. 7 as a function of weight percent ABS in the blend. As the ABS content is increased, the room temperature impact strength increases and the ductile–brittle transition temperature decreases. The ductile–brittle transition temperature is significantly reduced by compatibilization, however, the Izod impact strength is slightly lower. At

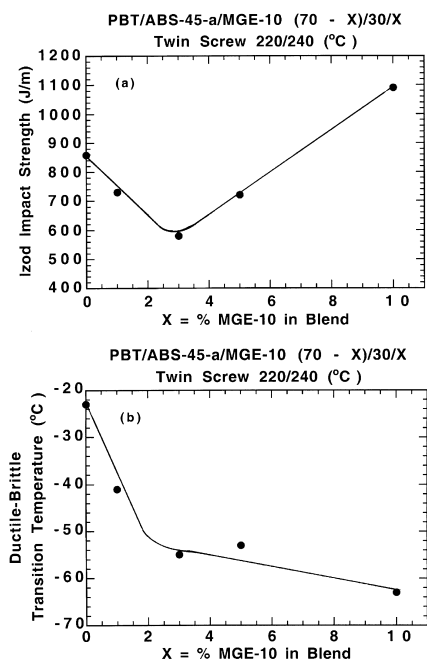


Fig. 13. Effect of MGE-10 content on room temperature notched Izod impact strength (a) and ductile–brittle transition temperature (b) of PBT/ABS-45-a/MGE-X (70-X)/30/X blends prepared in a twin screw extruder at 220°C and molded at 240°C.

least 30% ABS is required to significantly decrease the ductile–brittle transition temperature; higher ABS levels beyond this leads to only a minimal reduction in the ductile–brittle transition temperature. Fig. 8 shows the morphology of blends containing 10% ABS-45-a, with and without compatibilizer. The ABS domains of the uncompatibilized blend are large and contain many rubber particles. The blend containing 5% MGE-10, however, has a finer dispersion, but nevertheless it is apparent from Fig. 7 that 10% ABS is insufficient for toughening.

The rubber content of the ABS phase was varied by diluting ABS-45-a with SAN in the single screw extruder at 220°C. These ABS materials with reduced rubber content were then cryoground to a powder prior to mixing with PBT in the twin screw extruder. Fig. 9 shows the effect of rubber concentration in the ABS phase on the impact properties of ternary blends containing 30% ABS prepared in the twin screw extruder. At least 36% rubber in the ABS phase (10.8% rubber in the blend) is required to achieve a significant increase in the Izod impact strength and a significant reduction of the ductile–brittle transition temperature. At 27% rubber in the ABS, the impact strength of the compatibilized blend is higher than that of the uncompatibilized blend. The morphology of blends, with and without MGE-10, where the ABS-45-a was diluted with SAN to 18% rubber is shown in Fig. 10. The uncompatibilized blend reveals a poor ABS dispersion while the addition of 5% MGE-10 significantly disperses the ABS throughout the PBT matrix.

3.4. GMA content in the blend

Addition of the MGE-10 terpolymer to PBT/ABS blends was shown to produce a uniform dispersion of ABS that is more stable against coalescence during the molding cycle. To further examine the effect of this compatibilizer on blend properties, two series of experiments that vary the epoxide content in the blend, while keeping the ABS content fixed at 30%, were performed. The first series maintains a constant PBT/compatibilizer ratio in the blend while the GMA content in the compatibilizer is varied. In the second series, the GMA content of the compatibilizer is fixed while the PBT/compatibilizer ratio is varied.

Fig. 11 shows the Izod impact properties for a series of PBT/ABS-45-a/compatibilizer blends in the ratio of 65/30/5 prepared in the twin screw extruder for the case where the compatibilizer contains a varying amount of GMA. As the GMA content of the compatibilizer is increased, the room temperature Izod impact strength initially decreases reaching a minimum of 570 J/m at 5% GMA and then increases to 1100 J/m at a GMA content of 20%. However, the ductile–brittle transition temperature continually decreases from 17°C to below -50°C as the GMA content of the terpolymer is increased to 5%; any increase in GMA content beyond this point appears to have little effect. Fig. 12 shows the TEM photomicrographs for this series of blends. For blends where the compatibilizer contains 3% GMA or less, the ABS domains are not well dispersed. Increasing the GMA content to 5% provides uniform ABS dispersion throughout the matrix. Further increase in the GMA content to 20% leads to relatively little change in blend morphology.

Fig. 13 shows the impact strength of PBT/ABS-45-a/MGE-10 blends where the PBT/MGE-10 ratio is varied. As the compatibilizer content is increased, the room temperature Izod impact strength initially decreases reaching a minimum of 580 J/m at 3% MGE-10 and then increases to 1090 J/m at 10% MGE-10. The minimum impact strength seen here occurs when the overall GMA content in the blend is 0.3%. This behavior is similar to that observed when varying the GMA content of the MGE terpolymer (Fig. 11(a)), where the minimum impact strength occurs at nearly the same overall GMA content in the blend (0.25%). The ductile–brittle transition temperature decreases to below -50°C at an MGE-10 content of 3%; further increase in compatibilizer content beyond this has only a small effect on the ductile–brittle transition temperature. Fig. 14 shows TEM photomicrographs for this series of blends. The blend containing only 1% MGE-10 has a poor ABS dispersion compared to blends containing 3% MGE-10 or more.

Table 5 shows the tensile properties for the two series of blends described above. The presence of 30% ABS reduces the tensile modulus and yield strength relative to pure PBT. Addition of compatibilizer appears to have only a small effect on the tensile properties of these blends. There is, however, a general decrease in elongation at break as the

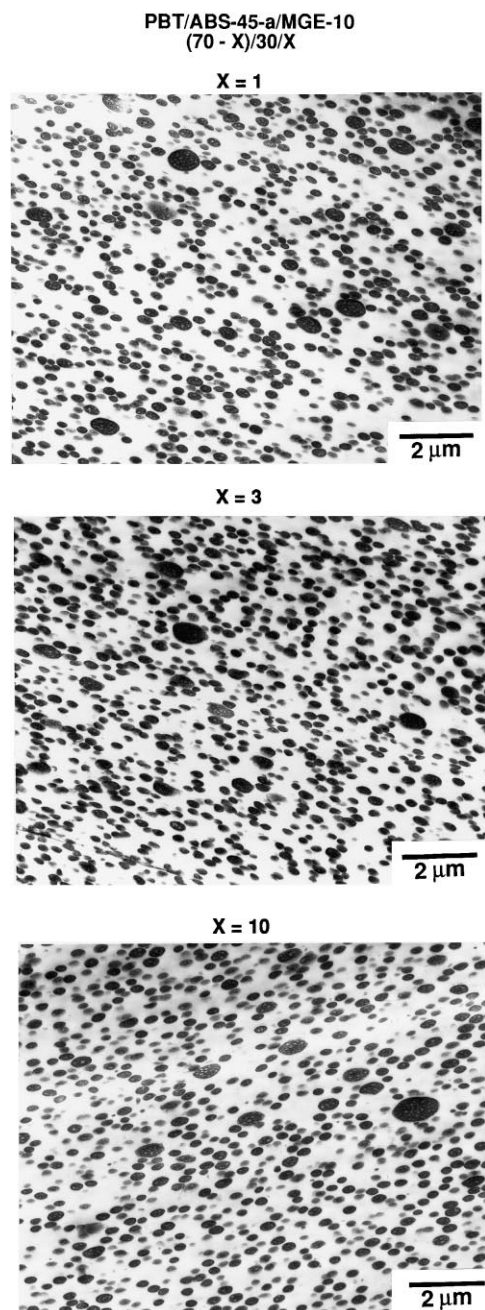


Fig. 14. TEM photomicrographs of PBT/ABS-45-a/MGE-10 (70-X)/30/X blends prepared in a twin screw extruder at 220°C and molded at 240°C. The rubber in the ABS phase is stained dark by OsO₄.

GMA content in the blend is increased, although the elongation at break is very high for the PBT/ABS-45-a/MGE-20 blend.

4. Conclusion

Very tough blends of PBT with various ABS materials can be achieved without compatibilizer within limited melt

processing situations. However, the morphology of these binary blends is unstable and phase coarsening can occur as demonstrated by TEM; this has a deleterious effect on blend properties. Terpolymers of MMA, GMA, and EA (MGE) were shown to be effective reactive compatibilizers for PBT/ABS blends providing improved low temperature impact properties, a finer dispersion of ABS domains, morphological stability, and a broadening of the processing window. The twin screw extruder is more effective for processing these reactive blends than the single screw extruder used here.

All ABS materials used were suitable for toughening PBT, however, ABS materials with higher rubber content and lower melt viscosity are more beneficial for producing tough uncompatibilized blends. The material designated as ABS-45-a has a high rubber content but only a moderate melt viscosity and was found to be superior for producing toughened blends. At least 30% ABS-45-a containing a minimum rubber content of 36% is required to significantly improve impact properties of these blends. Addition of the reactive compatibilizer improves ABS dispersion and lowers the ductile–brittle transition temperature for blends based on all ABS types.

Moderate amounts of GMA functionality in the compatibilizer (>5%) and small amounts of compatibilizer in the blend (<5%) significantly reduce the ductile to brittle transition temperature and improve ABS dispersion. While higher amounts of GMA functionality increase room temperature impact strength but produce only a minimal improvement in low temperature impact properties and increase blend viscosity which is undesirable for some processing techniques.

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References

- [1] Hage E, Hale W, Keskkula H, Paul DR. *Polymer* 1997;38:3237.
- [2] Hale W, Keskkula H, Paul DR, submitted to *Polymer*.
- [3] Hale W, Keskkula H, Paul DR, submitted to *Polymer*.
- [4] Utracki LA. *Polym. Eng Sci* 1995;35:2.
- [5] Lee P-C, Kuo W-F, Chang F-C. *Polymer* 1994;35:5641.
- [6] van Berkel RWM, van Hartingsveldt EAA, van der Sluijs CL. Polybutylene terephthalate. In: Olabisi O, editor. *Handbook of thermoplastics*. New York: Marcel Dekker, 1997:465.
- [7] Okamoto MSY, Kojima T, Inoue T. *Polymer* 1993;34:4868.
- [8] Hourston DJ, Lane S. In: Collyer AA, editor. *Rubber toughened engineering plastics*. London: Chapman and Hall, 1994:243.
- [9] Flexman EA. *Amer Chem Soc Advances in Chem. Ser.* 1993;233:79.
- [10] Cruz CA, Havriliak SJ, Slavin S. In *Proceedings of Additives'95: Advances in Additives and Modifiers for Polymers and Blends*,

- Executive Conference Management, 1995, Clearwater Beach, FL 22–24 Feb. 1995.
- [11] Benson CM, Burford RP. *J Mater Sci* 1995;30:573.
- [12] Kanai H, Auerbach A, Sullivan V. *Soc. Plast. Eng., ANTEC* 1994:2817.
- [13] Kanai H, Sullivan V, Auerbach A. *J Appl Polym Sci* 1994;53:527.
- [14] Hourston DJ, Lane S, Zhang H-X. *Polymer* 1991;32:2215.
- [15] Hourston DJ, Lane S, Zhang HX, Bootsma JPC, Koetsier DW. *Polymer* 1991;32:1140.
- [16] Moffett AJ, Dekkers MEJ. *Polym Eng Sci* 1992;32:1.
- [17] Binsack R, Rempel D, Humme G, Ott K-H. (Bayer) US Pat. 4 292 233, 1981.
- [18] Binsack R, Rempel D, Lindner C, Morbitzer L, (Bayer) US Pat. 4 535 124, 1985.
- [19] Lindner C, Binsack R, Rempel D, Ott K-H. (Bayer) US Pat. 4 417 026, 1983.
- [20] Wang I-CW, (General Electric) US Pat. 4 753 986, 1988.
- [21] Paul DR. In: Paul DR, Newman S, editors. *Polymer blends*. New York: Academic Press, 1978, Ch. 1, Vol. 1, p. 2.
- [22] Paul DR. In: Paul DR, Newman S, editors. *Polymer blends*. New York: Academic Press, 1978, Ch. 12, Vol. 2, p. 35.
- [23] Xanthos M, Dagli SS. *Polym. Eng. Sci.* 1991;31:929.
- [24] Brown SBA. Survey of chemical reactions of monomers and polymers during extrusion process. In: Xanthos M, editor. *Reactive extrusion: principles and practice*. New York: Oxford Press, 1992: 78.
- [25] Bucknall CB. *Toughened plastics*. Essex, England: Applied Science Publishers Ltd., 1977.
- [26] Brady AJ, Keskkula H, Paul DR. *Polymer* 1994;35:3665.
- [27] Paul DR. *Pure and Appl Chem* 1995;67:977.
- [28] Stewart ME, George SE, Miller RL, Paul DR. *Polym Eng Sci* 1993;33:675.
- [29] Hert M. *Angew Makromol Chem* 1992;196:89.
- [30] Hourston DJ, Lane S, Zhang HX. *Polymer* 1995;36:3051.
- [31] Laurienzo P, Malinconico M, Martuscelli E, Volpe G. *Polymer* 1989;30:835.
- [32] Lombardo BS, Keskkula H, Paul DR. *J Appl Polym Sci* 1994;54:1697.