Polymer Blends of PBT and PP Compatibilized by Ethylene-co-glycidyl Methacrylate Copolymers

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SYNOPSIS

The ethylene-co-glycidyl methacrylate (EG) copolymer is an efficient reactive compatibilizer for polymer blends of poly(butylene terephthalate) (PBT) and polypropylene (PP). During melt processing, the epoxy functional group of the EG copolymer can react with the PBT carboxylic acid and/or hydroxyl terminal groups at the interface to form various EG-g-PBT copolymers. These in situ formed grafted copolymers tend to concentrate along the interface to reduce the interfacial tension at the melt and result in finer phase domains. Higher glycidyl methacrylate (GMA) content in the EG copolymer or a higher quantity of the EG compatibilizer in the blend results in a better compatibilized blend in terms of finer phase domains, higher viscosity, and better mechanical properties. The presence of only 50 ppm catalyst (ethyltriphenyl phosphonium bromide) in the EG compatibilized blend further improves the blend compatibility substantially. © 1996 John Wiley & Sons, Inc.

INTRODUCTION

Straight blends between polyalkyl terephthalates and polyolefins are considered to be immiscible and incompatible. Only a very limited number of studies on these blends have been reported; however, they have received considerable interest lately.[1–12] Rudin et al.[1] investigated oriented monofilaments from blends of poly(ethylene terephthalate) (PET) and polypropylene (PP). Yang and Smith[2] studied the melting and solidification behavior of poly(butylene terephthalate) (PBT) and high density polyethylene (HDPE). Barlow and coworkers[3,4] found that the addition of a small amount of the triblock copolymer of styrene–butene–1–styrene (SEBS, Kraton G1652 from Shell Chemical Company) in PET/HDPE blends resulted in greatly improved ductility of these incompatible blends. The compatibilization of these blends is not the same as a classic compatibilizer by the incorporation of segments (of the block copolymer) into the two incompatible blend components to form molecular bridges across the interface. Bataille et al.[5] used a copolymer of PP and polyacrylic acid to compatibilize PET/PP blends that resulted

in higher yield stress and better phase dispersion. Xanthos and colleagues[6] used an acrylic acid functionalized PP as compatibilizer in PET/PP blends that resulted in finer dispersed phase morphology and improved mechanical properties. The nature of compatibilization is that of specific interactions rather than a true covalent reaction.[6] Cecere et al.[7] studied rubber toughened PBT by using maleic anhydride (MAH) grafted ethylene–propylene rubber (EPR). The MAH functional groups can react with PBT hydroxyl terminal groups to form EPR-g-PBT copolymer. Hourston and cohorts[8] used a maleated PBT as a reactive compatibilizer for the blends of PBT with ethylene–propylene–diene monomer (EPDM) and polybutadiene (PB) rubbers. Model compound experiments indicated that the electron-deficient maleate groups of the modified polyester can react with the unsaturated groups in these unsaturated rubbers via an ene mechanism.[8] Akkapeddi and Van Buskirk[9] used ethylene-co-glycidyl methacrylate (EG) as a reactive compatibilizer for blends of PET with various polyolefins through covalent reaction between the epoxy groups in the EG and carboxylic acid groups in the PET to form the EG-g-PET copolymer. Sambarn and Jabarin[10] used a polyolefin-g-MAH copolymer as a reactive compatibilizer for the PET/HDPE blends. Dagli and Kamdar[11] studied the effects of component addition
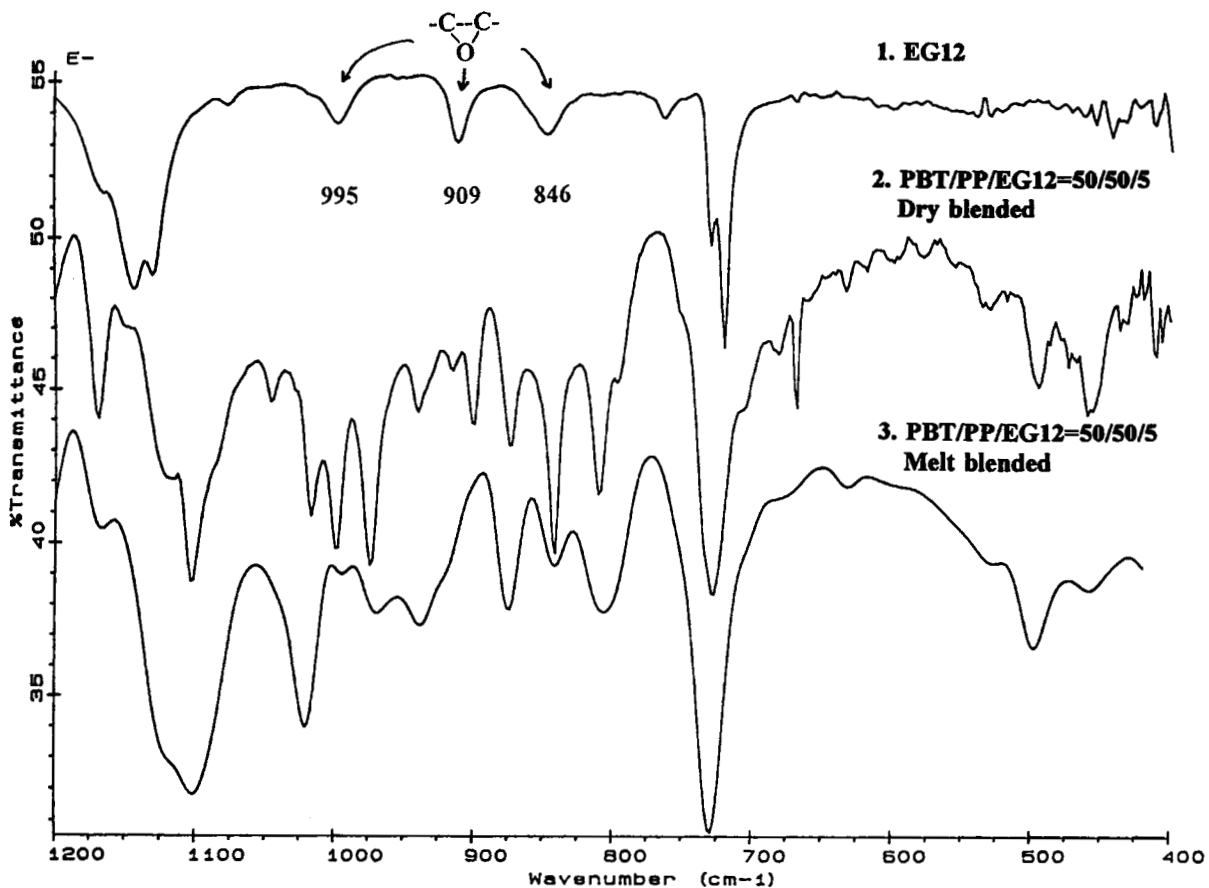
Figure 1   FTIR spectra of EG12 and PBT/PP/EG12, before and after melt blending.
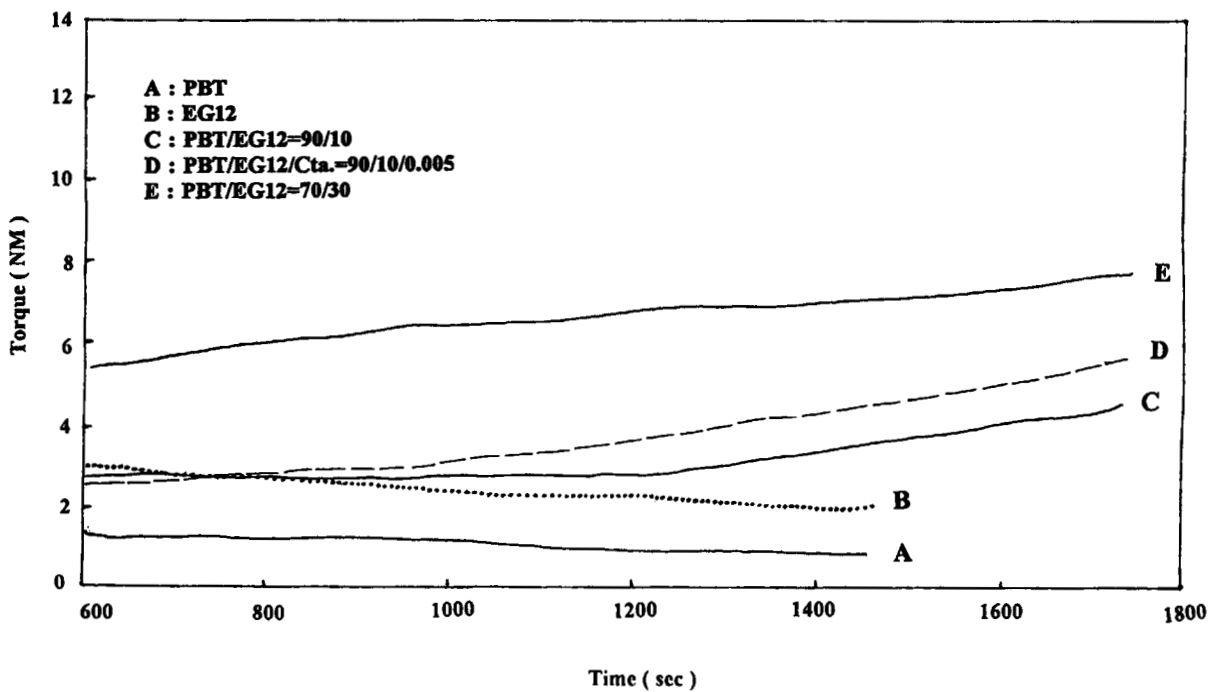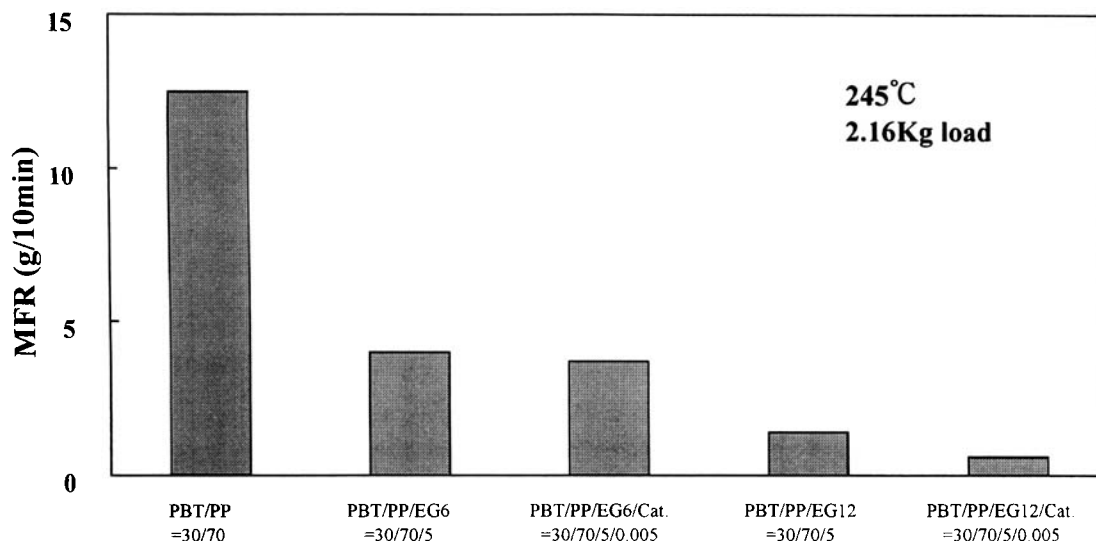

Figure 2   Torque versus time curves for PBT, EG12, and various PBT/EG12 blends.
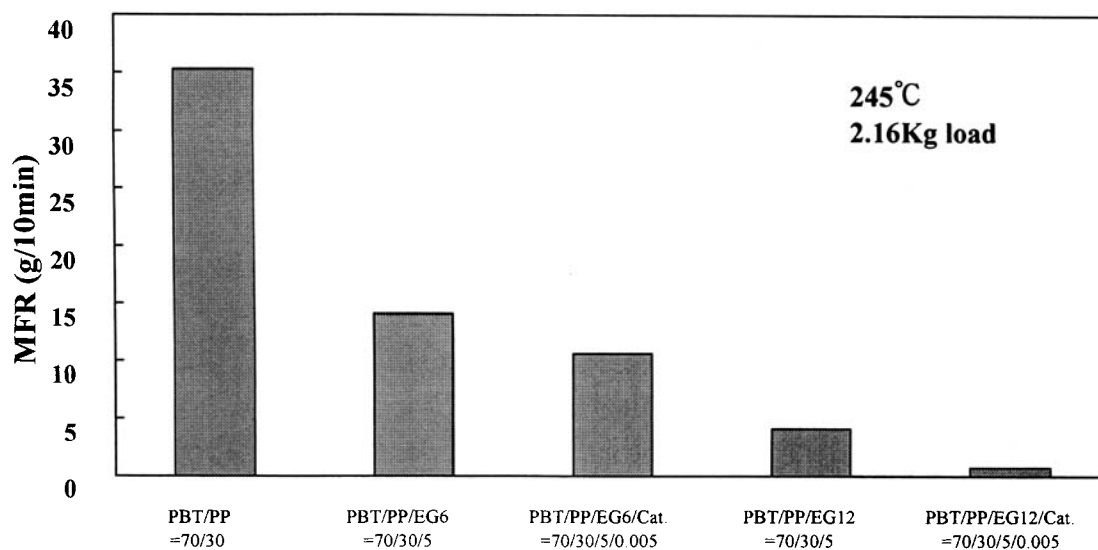
Figure 3 Effect of EG and catalyst on melt flow rate of the PP-dominant PBT/PP blends.

protocol on reactive compatibilization of PET/ HDPE blends. Boutevin et al.[12] investigated the compatibilization of PBT/PP blends by using a specially synthesized polyisoprene-*b*-PBT copolymer that resulted in significant reduction in particle size and increase of tensile elongation. Holsti–Miettinen and coworkers[13] used the epoxy functionalized polymer, ethylene/ethyl acrylate/glycidyl methacrylate (GMA), as reactive compatibilizer for PBT/ PP and LCP/PP blends.

In a recent review on compatibilization of thermoplastics blends,[14] we mentioned that polyester-related reactive compatibilized blends are currently

the second largest group after the polyamide-related blends. Graft or block copolymers, suitably functioning as compatibilizers for immiscible blends, can be formed *in situ* through chemical bonding, ionic or covalent, during melting processes. Typically, a C-X reactive copolymer (X is the reactive groups which can be a terminal group or randomly distributed in the main chain) can act as a compatibilizer for the A/B blend, provided that C is identical or miscible with component A while X is able to react with component B to form the C-X-B graft or block copolymer. Reactive compatibilization is not universally applicable to all polymer blends, only to

Figure 4 Effect of EG and catalyst on melt flow rate of the PBT-dominant PBT/PP blends.

RHEOLOGY AT 240 °C
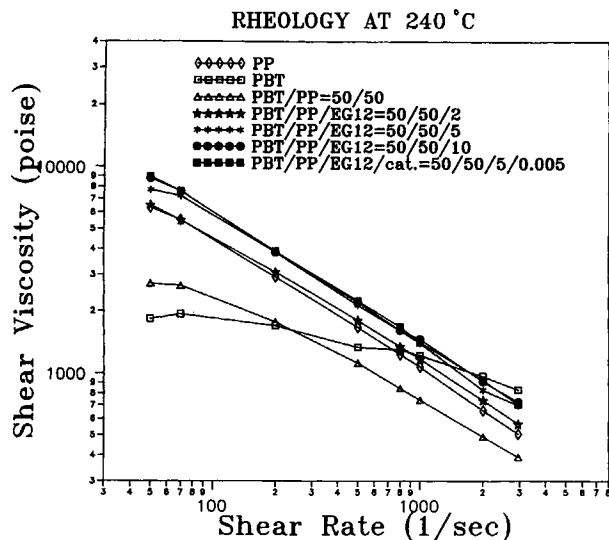
Figure 5   Shear viscosity versus shear rate curves of PBT, PP, and various uncompatibilized and compatibilized 50/50 PBT/PP blends.

those pairs having at least one of the blend constituents possessing certain functional groups that can react with the reactive compatibilizer. Most polyesters inherently contain carboxylic acid and/or hydroxyl terminal groups that make them particularly suitable as one blend component for reactive compatibilization. In most of the above-mentioned literature on polyalkyl terephthalate/polyolefin blends, they were compatibilized by copolymers containing an epoxy, MAH, or acrylic acid functional group. The epoxy-containing copolymers are probably the best candidates as reactive compatibilizers for polyester-related blends because the reactions between epoxy and carboxylic acid or hydroxyl groups are well-known chemistry.

A series of continuous investigations on *in situ* compatibilization of various blends using epoxy-containing copolymers or polymers as compatibilizers was carried out in this lab.[15-24] This article reports on details of the PBT/PP blends compatibilized by EG copolymers.

EXPERIMENTAL

PPs with various melt flow rates (MFRs) were obtained from Taiwan Polypropylene Company. PBT D-201 natural grade is the product of Sinkong Synthetic Fibers Corporation of Taiwan. EG copolymers, Igetabond 2C (6% GMA) and Igetabond E (12% GMA), were purchased from Sumitomo Chemical Company of Japan. Ethyltriphenyl phos-

phonium bromide used as catalyst was purchased from Merck Company.

Melt blending was carried out using a 30-mm corotating twin-screw extruder with L/D of 36 and barrel temperature ranging from 245 to 255°C. The extruded pellets were dried and injection molded into standard $\frac{1}{8}$-in. ASTM tensile and impact testing specimens using an Arburg 3-oz. injection molding machine.

Capillary rheological measurements were performed on a capillary rheometer (L/D 40, orifice radius 0.02 in., orifice length 0.8 in.) from Kayeness Company model Galaxy X at 240°C. To verify the reaction between epoxy and PBT based on the viscosity increase, 35 g of sample were tested at 260°C and 30 rpm in a Brabender Plastic-Corder.

Fourier transform infrared spectroscopic (FTIR) analysis to detect epoxy groups was performed on a Nicolet 500 Infrared Spectrophotometer.
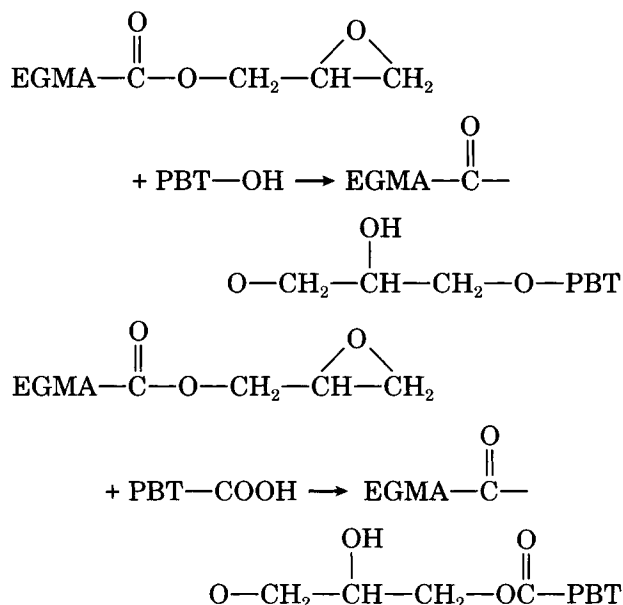
Morphologies of the cryogenically fractured surfaces were etched to remove the minor component, then examined by scanning electron microscopy (SEM, Model S-570, Hitachi Co. of Japan). Etching out of the PP phase of the blends was carried out by reflux heated xylene solvent for 1 h. Mixed solvent of phenol/chloroform ($\frac{2}{3}$) was used to etch out the PBT phase of the blends. All SEM morphologies were taken in a region midpoint between the central line and skin of the injection molded specimens on the plane perpendicular to the injection flow direction. Microtomed thin sections of the injection molded specimens perpendicular to the flow direction were stained in ruthenium tetraoxide (RuO_4) solution prior to transmission electron microscopy (TEM, JEM-100S, Jeol-Technics Co., Japan) examination.

Unnotched Izod impact tests were carried out at ambient conditions. Standard tensile tests were also carried out at ambient conditions using an extensometer with a crosshead speed of 50 mm/min according ASTM-D638. The impact critical strain energy release rates (G_c) were determined by varying the depth of the notch (5 mil) according to the previously developed method.[25]
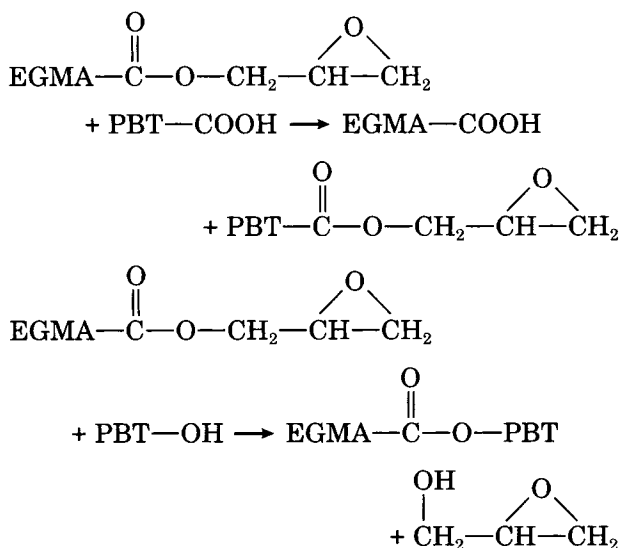
RESULTS AND DISCUSSION

Chemistry

The major chemical reactions involved in this reactive compatibilized blend system are from the epoxy groups in the EG copolymer with PBT terminal carboxylic acid and/or hydroxyl groups and can be expressed by the following reactions:

$$EGMA-\overset{\overset{O}{\|}}{C}-O-CH_2-\overset{\diagup O\diagdown}{CH}-CH_2$$

$$+ \; PBT-OH \longrightarrow EGMA-\overset{\overset{O}{\|}}{C}-$$

$$\overset{\overset{OH}{|}}{O-CH_2-CH-CH_2-O-PBT}$$

$$EGMA-\overset{\overset{O}{\|}}{C}-O-CH_2-\overset{\diagup O\diagdown}{CH}-CH_2$$

$$+ \; PBT-COOH \longrightarrow EGMA-\overset{\overset{O}{\|}}{C}-$$

$$\overset{OH}{\underset{|}{O-CH_2-CH-CH_2}}-\overset{\overset{O}{\|}}{OC}-PBT$$

Ethyltriphenyl phosphonium bromide catalyzes the above two reactions and the possible catalytic mechanisms were described in our previous article.[23]

Transesterification reactions can also take place between the acrylate ester group and the terminal —OH and —COOH groups of polyesters reported previously.[26-28] Therefore, some degree of similar transesterification may also take place in this system as shown by the following reactions:

$$EGMA-\overset{\overset{O}{\|}}{C}-O-CH_2-\overset{\diagup O\diagdown}{CH}-CH_2$$

$$+ \; PBT-COOH \longrightarrow EGMA-COOH$$

$$+ \; PBT-\overset{\overset{O}{\|}}{C}-O-CH_2-\overset{\diagup O\diagdown}{CH}-CH_2$$

$$EGMA-\overset{\overset{O}{\|}}{C}-O-CH_2-\overset{\diagup O\diagdown}{CH}-CH_2$$

$$+ \; PBT-OH \longrightarrow EGMA-\overset{\overset{O}{\|}}{C}-O-PBT$$

$$+ \; \overset{OH}{\underset{|}{CH_2}}-\overset{\diagup O\diagdown}{CH}-CH_2$$

These transesterification reactions are believed to be much slower than the epoxy coupling reactions and can be neglected unless under an extended period time of melting. These transesterifications are not expected to cause overall molecular weight and viscosity increases of the blends.

FTIR

The IR peaks at 846, 909, and 995 cm^{-1} are characteristic of epoxy groups in EG as shown in curve 1 of Figure 1. Curves 2 and 3 of Figure 1 compare the IR spectra of the 50/50/5 PBT/PP/EG12 blend before and after melt blending. The observed disappearance of peaks 909 and 995 cm^{-1} after melt blending indicates epoxy ring-opening reactions indeed occur, but we were unable to positively identify the chemical structures of the reaction products from the spectra.

Torque Versus Time

Figure 2 compares the torque versus time curves of PBT, EG12, and various PBT/EG12 mixtures during melt mixing by omitting the front unstable region ($t < 600$ s). It clearly shows that PBT and EG12 individually have lower torque values than their mixtures. The torque of the 90/10 PBT/EG12 mixture (curve C) is significantly higher than the additive average of the blend components (curves A and B). The additional 50-ppm catalyst in the blend results in a further increase of the torque of the blend (compare curves C and D). The mixture with higher epoxy content, 70/30 PBT/EG12, has the highest torque (curve E). The viscosity increase (torque increase) can be attributed to the overall molecular weight increase from the anticipated epoxy coupling reactions to form EG-g-PBT copolymers. As mentioned earlier, transesterification reactions between EG and PBT are not expected to cause a viscosity increase.

MFRs

Figure 3 clearly demonstrates the effect of epoxy content and the presence of catalyst on the resulting viscosities (in terms of MFRs) of the PP-dominant 30/70 PBT/PP blends. Figure 4 shows a trend similar to Figure 3 for the PBT-dominant 70/30 PBT/PP blends. Because EG copolymers are structurally similar to PP, they are expected to be more compatible with PP than with PBT. Additionally, PP has a lower melting temperature than PBT. Therefore, EG is expected to reside mainly in the PP phase during the earlier stages of melt blending while the later coupling reactions between epoxy groups of EG and PBT terminal groups should take place along the interface between PP and PBT. As soon as the EG-g-PBT copolymers are formed *in situ,* they tend to anchor at the interface to reduce interfacial tension and function as phase emulsifiers of the blend.
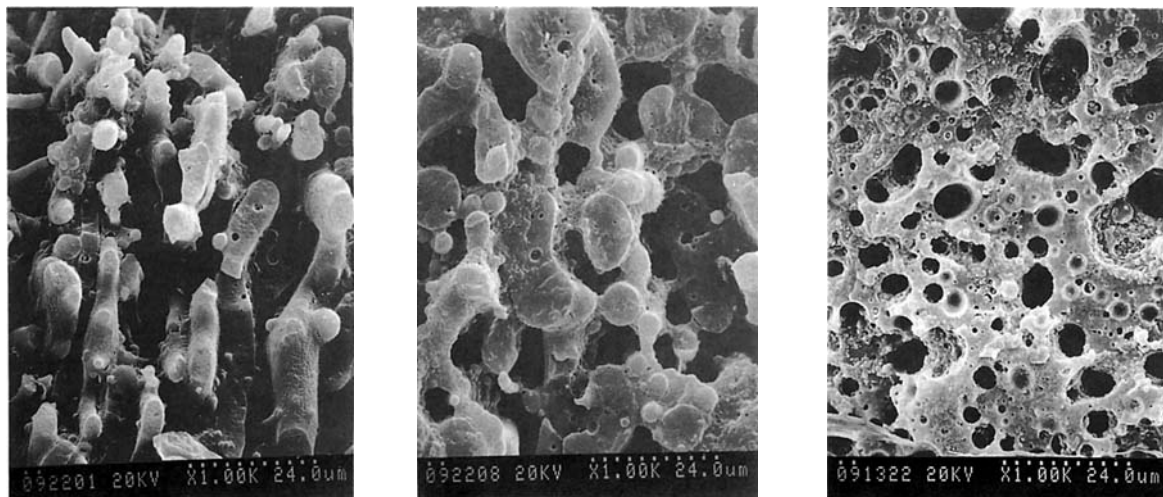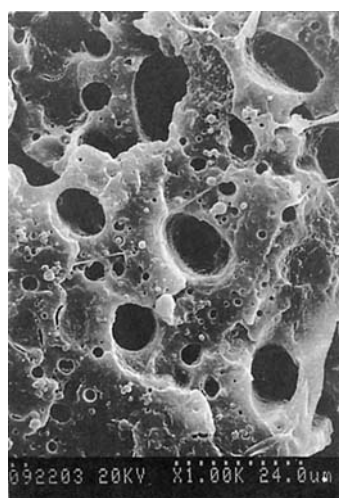
A. PBT/PP=50/50,   B. PBT/PP/EG12=50/50/2,   C. PBT/PP/EG12=50/50/5
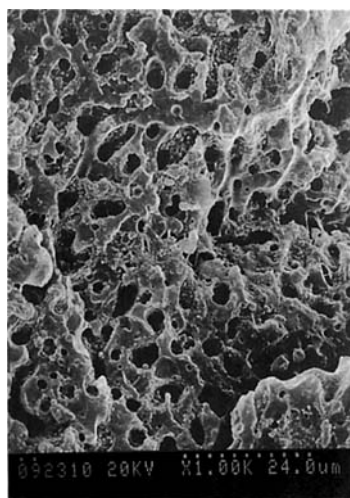
D. PBT/PP/EG12=50/50/10,   E. PBT/PP/EG6=50/50/5.

Figure 6   SEM micrographs of the uncompatibilized and compatibilized 50/50 PBT/PP blends: (A) 50/50 PBT/PP, (B) 50/50/2 PBT/PP/EG12, (C) 50/50/5 PBT/PP/EG12, (D) 50/50/10 PBT/PP/EG12, and (E) 50/50/5 PBT/PP/EG6.

Capillary Rheometry

Figure 5 gives the shear rate versus viscosity curves of PBT, PP, uncompatibilized, and various compatibilized 50/50 PBT/PP blends. Due to the expected more rigid chain structure of PBT than PP, PBT shows significantly less shear thinning than does PP. Essentially all the blends in this component ratio (50/50) show similar shear thinning behavior as the PP component. The uncompatibilized blend has the lowest viscosity, even lower than both blend constituents at high shear rates. The viscosity of the blend increases with increasing quantity of the

EG12 compatibilizer up to 5 phr. Blends containing EG12 compatibilizer higher than 5 phr, or with the presence of catalyst, show no further viscosity increase. The general trend from this capillary rheometrical study is very similar to that from previous MFR data.

SEM Morphologies

Figure 6 (A–F) shows the SEM micrographs of the uncompatibilized and compatibilized 50/50 PBT/PP blends where the PBT component has been re-
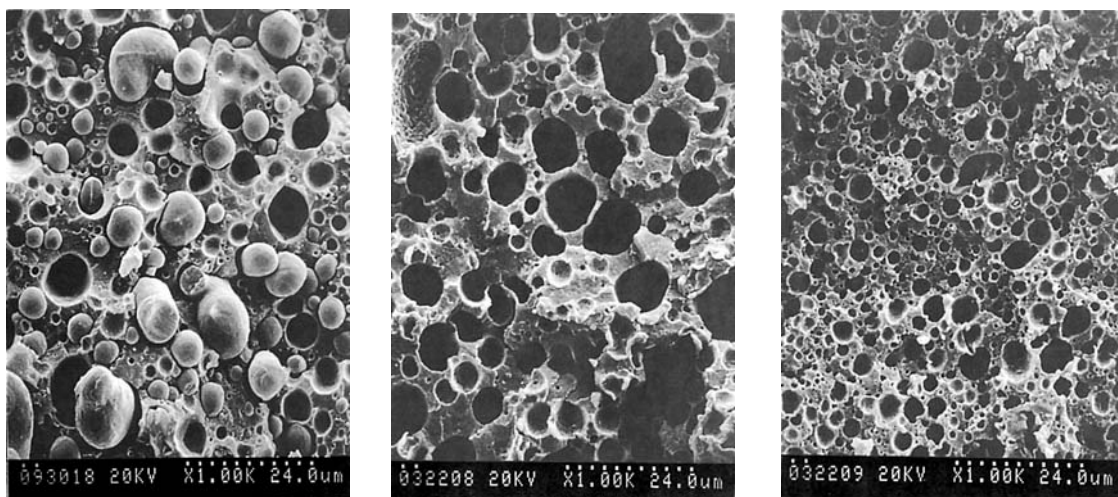
A. PBT/PP=70/30 (non-etched),   B. PBT/PP=70/30 (Etched)   C. PBT/PP/EG6=70/30/5 (Etched),
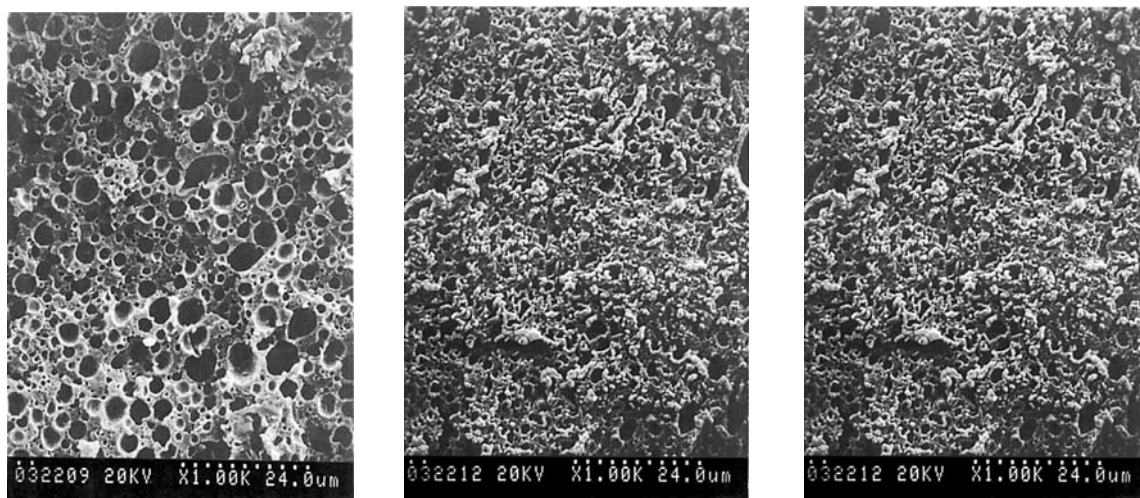
D. PBT/PP/EG12=70/30/5 (Etched),   E. PBT/PP/EG12/Cat.=70/30/5/0.005 (Etched),   F. PBT/PP/EG12/Cat. =70/30/5/0.005 (Non-etched

Figure 7   SEM micrographs of the uncompatibilized and compatibilized 70/30 PBT/PP blends: (A) 70/30 PBT/PP (nonetched), (B) 70/30 PBT/PP (etched), (C) 70/30/5 PBT/PP/EG6 (etched), (D) 70/30/5 PBT/PP/EG12 (etched), (E) 70/30/5/0.005 PBT/PP/EG12/Cat. (etched), and (F) 70/30/5/0.005 PBT/PP/EG12/Cat. (nonetched).

moved by solvent etching. Figure 6(A) shows the cocontinuous structure of the uncompatibilized 50/50 PBT/PP blend. Figure 6(B–D) shows the morphologies of the blends containing various amounts of EG12 compatibilizer, from 2 to 10 phrs. The domain size decreases with the increase of compatibilizer quantity as would be expected from any efficient compatibilizer. The estimated volume fraction of the PP phase in the blends increases by increasing the quantity of the added compatibilizer but such volume increase is much more than by simple addition of PP and compatibilizer. Later TEM micrographs show that a significant amount of PP or EG-g-PBT distributed in the PBT phase from these compatibilized blends may result in incomplete solvent etching. The increase of the compatibilizer content also has the tendency to shift the cocontinuous structure of the compatibilized blends at a lower level of EG12 [2 phr, Fig. 6(B)] into discontinuous morphology with PBT as the dispersed particles [Fig. 6(C, D)]. Figure 6(E) is the SEM micrograph of the blend containing 5 phr of EG6 com-
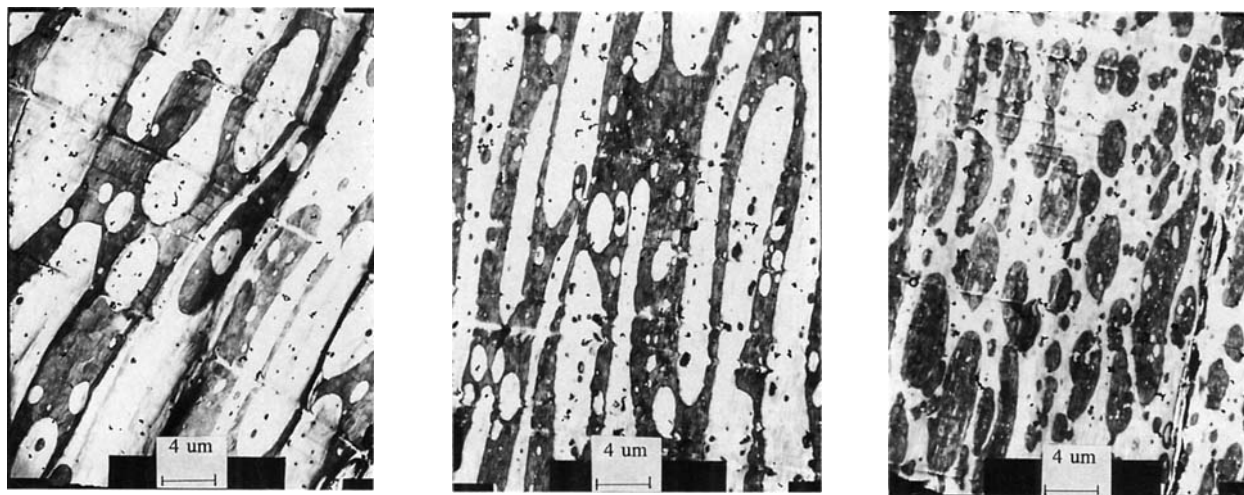
A. PBT/PP=50/50, B. PBT/PP/EG12=50/50/5, C. PBT/PP/EG12/Cat.=50/50/5/0.005.

Figure 8 TEM micrographs of the uncompatibilized and compatibilized 50/50 PBT/PP blends, (A) 50/50 PBT/PP, (B) 50/50/5 PBT/PP/EG12, and (C) 50/50/5/0.005 PBT/PP/EG12/Cat.

patibilizer where the average size of the PBT particle is larger than the corresponding blend containing 5 phr of EG12 compatibilizer [Fig. 6(C)].

Figure 7 demonstrates the effect of epoxy content in the compatibilizer and the presence of catalyst
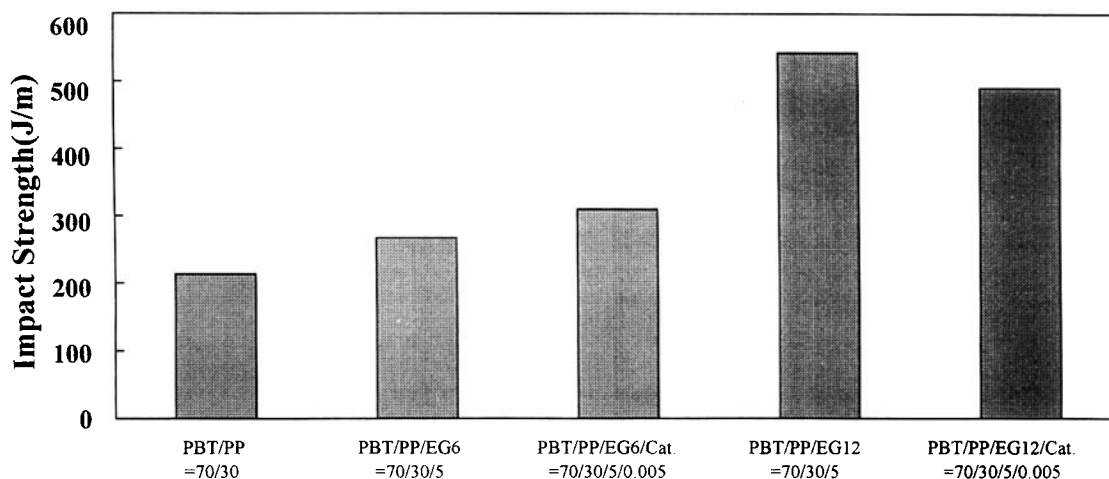
on the domain size of the dispersed PP particles. Figure 7(A, B) is the etched and nonetched fracture surface SEM micrographs of the uncompatibilized 70/30 PBT/PP blend. The nonetched SEM micrograph of this uncompatibilized blend [Fig. 7(A)]

Table I Mechanical Properties of PBT/PP Blends

	Unnotch (J/m)	Tensile Strength (MPa)	Tensile Elongation (%)	Gc (kJ/m^2)
70/30 PBT/PP	139	32.1	4.27	2.81
50/50 PBT/PP	183	31.3	6.04	2.36
30/70 PBT/PP	211	29.3	7.78	—
70/30/5 PBT/PP/EG6	262	34.3	5.34	4.39
70/30/5/0.005 PBT/PP/EG6/cat.	330	36.0	6.20	—
70/30/5 PBT/PP/EG12	381	38.2	10.85	4.92
70/30/5/0.005 PBT/PP/EG12/cat.	591	37.8	9.82	—
70/30/20 PBT/PP/EG12	648	34.5	16.40	—
50/50/2 PBT/PP/EG6	236	26.9	5.77	—
50/50/5 PBT/PP/EG6	251	29.8	6.86	6.35
50/50/10 PBT/PP/EG6	397	26.8	9.09	—
50/50/5/0.005 PBT/PP/EG6/cat.	532	31.0	7.91	—
50/50/5/0.02 PBT/PP/EG6/cat.	468	30.4	7.07	—
50/50/2 PBT/PP/EG12	359	30.0	8.72	—
50/50/5 PBT/PP/EG12	240	28.0	9.20	5.60
50/50/10 PBT/PP/EG12	400	27.1	11.29	—
50/50/20 PBT/PP/EG12	324	27.9	7.00	—
50/50/5/0.005 PBT/PP/EG12/cat.	526	32.8	13.84	—
50/50/5/0.02 PBT/PP/EG12/cat.	489	33.4	12.10	—
30/70/5 PBT/PP/EG6	294	30.4	8.32	—
30/70/5/0.005 PBT/PP/EG6/cat.	297	28.5	9.14	—
30/70/5 PBT/PP/EG12	381	29.1	9.92	—
30/70/5/0.005 PBT/PP/EG12/cat.	389	27.0	12.57	—

Izod Impact (Unnotched,25°C)

Figure 9 Effect of EG copolymer and catalyst on unnotched impact strength of the uncompatibilized and compatibilized 70/30 PBT/PP blends.

clearly shows poor interfacial adhesion between matrix PBT and the dispersed PP particles. Figure 7(C, D) shows that the PP particle sizes of the etched compatibilized blends (by EG6 and EG12) were reduced substantially compared with the uncompatibilized blend [Fig. 7(B)]. The EG12 compatibilizer was more efficient than EG6 in reducing the PP particle size [comparing Fig. 7(D) and (C)]. Figure 7(E) indicates that the presence of only 50 ppm catalyst results in further reduction of the dispersed PP particle size [comparing Fig. 7(E) and (D)]. Figure 7(F) shows the nonetched morphology from the same catalyzed blend as Figure 7(E) where

the phase contrast nearly disappears, indicating stronger interfacial adhesion of the blend.

TEM

TEM analysis can avoid the SEM problems caused by incomplete solvent etching on some compatibilized blends mentioned earlier. Figure 8 shows the TEM micrographs of the uncompatibilized and compatibilized 50/50 PBT/PP blends. All the TEM micrographs were taken from the plane perpendicular to the flow direction of injection molding. Figure 8(A) gives the uncompatibilized blend,

Tensile Elongation
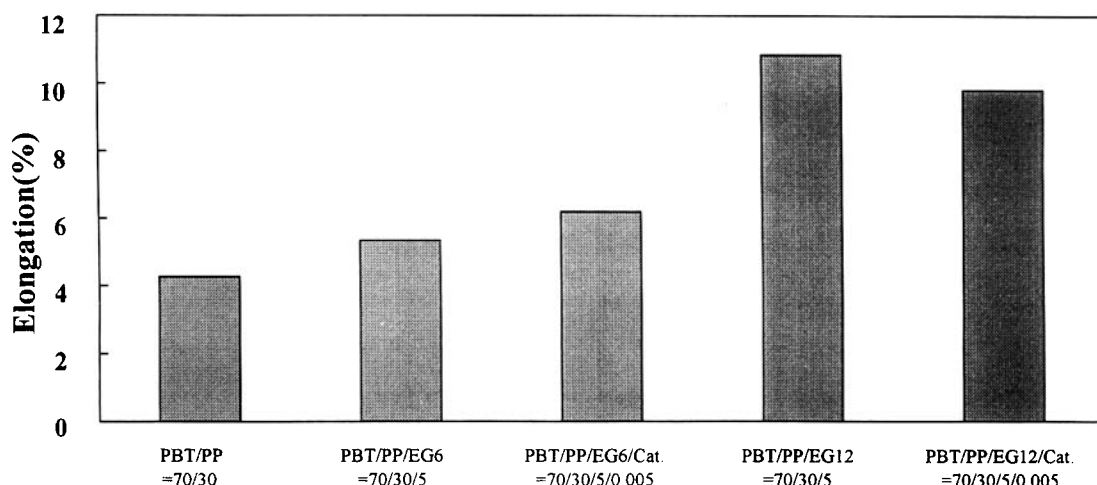
Figure 10 Effect of EG copolymer and catalyst on tensile elongation of the uncompatibilized and compatibilized 70/30 PBT/PP blends.
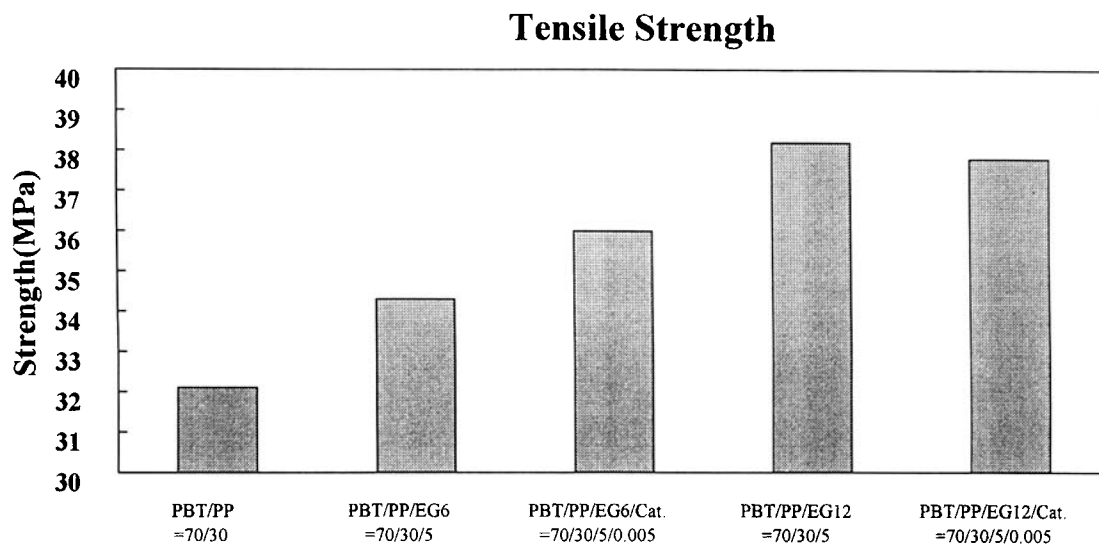
Tensile Strength

Figure 11   Effect of EG copolymer and catalyst on tensile strength of the uncompatibilized and compatibilized 70/30 PBT/PP blends.

showing coarser morphology with a relatively smoother surface on the PBT phase (darker phase). Figure 8(B) shows the domain size has been reduced substantially for the blend containing 5 phr EG12. The surface of the PBT phase in this blend is rather rough, which indicates the presence of PP or EG-g-PBT in the PBT phase. The PP phase contains some small PBT droplets. Figure 8(C) shows the catalyzed compatibilized blend where the PBT phase becomes the elongated dispersed phase. All three TEM micrographs have about equal volume fractions from the PBT and PP phases and such observations provide evidence

to support our previous suspicion of incomplete solvent etching from the SEM micrographs for some compatibilized blends [comparing Fig. 8(C) and Fig. 6(D)].

Mechanical Properties

Mechanical properties including unnotched Izod impact, tensile properties, and strain energy release rate (G_c) of the blends investigated are summarized in Table I. Because both PBT and PP are brittle under standard notched Izod impact testing, all the PBT/PP blends, uncompa-

Figure 12   SEM micrographs of the tensile fractured surfaces of the uncompatibilized and compatibilized 70/30 PBT/PP blends: (A) 70/30 PBT/PP, (B) 70/30/5 PBT/PP/EG6, and (C) 70/30/5 PBT/PP/EG12.

tibilized or compatibilized, are also brittle with nearly identical impact strength as would be expected (data not shown here). Unnotched impact strength is more appropriate to use to differentiate the toughness change of the notch sensitive blends through compatibilization. The unnotched impact strength increases by increasing the quantity of EG compatibilizer. The only exception found is the blend containing 20 phr EG12 (50/50/20 PBT/PP/EG12), for which the impact strength is actually decreased. Higher GMA content in the compatibilizer also results in higher impact strength (EG12 > EG6). The presence of merely 50 ppm of catalyst is able to improve impact toughness substantially. Increasing catalyst quantity to 200 ppm does not give further improvement in impact strength. Figure 9 gives only the 70/30 PBT/ PP series of blends to demonstrate the trend of impact strength improvement mentioned above; a similar trend also applies to other series of blends as shown in Table I.

Tensile elongation represents the tensile toughness of the material. The observed trend of the tensile elongation improvement due to compatibilization is almost identical to that of the unnotched impact. Figure 10 shows tensile elongations of the PBT-dominant 70/30 PBT/PP blends where the only exception is the slight drop found for the catalyzed blend. The trend on tensile strength of the PBT-dominant 70/30 PBT/PP blends shown in Figure 11 is identical to that in tensile elongation. The trends of tensile strength on the other two series of blends, 50/50 PBT/PP and 30/70 PBT/PP, are not very consistent as shown in Table I. Phase transition from cocontinuous to PBT as a dispersed phase in the 50/50 PBT/PP series and the rubbery nature of the EG copolymers are probably responsible for such inconsistent results observed in tensile strength. Figure 12 shows the SEM micrographs of the tensile fractured surfaces of the uncompatibilized and compatibilized 70/30 PBT/PP blends. The matrix (PBT) shear yielding of the uncompatibilized blend [Fig. 12(A)] is rather limited, an indication of poor interfacial adhesion. Extensive shear yielding occurs with the compatibilized blends and the extent of shear yielding increases with the increase of GMA content in the SG copolymer [comparing SG6 of Fig. 12(B) with SG12 of Fig. 12(C)]. These SEM morphologies from the tensile fractured surfaces support the improvement of tensile elongation and tensile strength of the blends due to better compatibilization.

Data on blends, selected to determine their strain energy release rates (G_c) using the Izod impact approach by varying their notch depths, are also sum-marized in Table I. A G_c increase of 2–3 times for the compatibilized blends over the uncompatibilized counterparts was observed.

CONCLUSIONS

The EG copolymer was demonstrated to be an effective in situ reactive compatibilizer for immiscible and incompatible PBT/PP blends. The epoxy groups of EG copolymer can have a covalent reaction with the PBT terminal carboxylic acid and/or the terminal hydroxyl groups at their interface to form various EG-g-PBT copolymers. These in situ formed grafted copolymers tend to reside along the interface to reduce the interfacial tension in the melt and result in finer domains. Additionally, the interfacial adhesion is also expected to be increased and results in better mechanical properties. Higher GMA content in EG or higher quantity of EG in the blends results in finer phase domains, higher viscosity, and better mechanical properties. The presence of as little as 50 ppm catalyst is able to further improve the compatibility of the compatibilized blends.

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