

Reactive Compatibilization of PET and PPE Blends by Epoxy Couplers

DA-WEI LO, CHIH-RONG CHIANG, FENG-CHIH CHANG

Institute of Applied Chemistry, National Chiao-Tung University, Hsinchu, Taiwan, Republic of China

Received 25 July 1996; accepted 9 January 1997

ABSTRACT: A commercially available tetrafunctional epoxy monomer was demonstrated to be an effective reactive compatibilizer for blends of poly(ethylene terephthalate) (PET) and polyphenylene ether (PPE). It requires about 10–20% by weight of a conventional reactive compatibilizer to achieve the same level of compatibilization in terms of domain size reduction and mechanical property improvement. This epoxy monomer is able to contact and react with PET and PPE simultaneously during high shear extruder blending to form the desirable PET–epoxy–PPE mixed copolymer. This mixed copolymer contains PET and PPE segments that tend to anchor at the interface to act as an efficient compatibilizer of the PET and PPE blends. © 1997 John Wiley & Sons, Inc. *J Appl Polym Sci* **65**: 739–753, 1997

Key words: polymer blend; reactive compatibilizer; coupling agent; PPE; PET; epoxy

INTRODUCTION

The amount of yearly published literature on the area of nonreactive compatibilization has maintained a steady growth rate during last two decades. The amount of literature on reactive compatibilization was relatively scarce before 1990, but has increased at an exponential rate since then. Reactive compatibilization of polymer blends has been the subject of a few recent reviews.^{1–3}

A conventional reactive compatibilization of a polymer blend A/B is a graft or block reactive C–X copolymer provided that C is structurally similar or miscible with component A and X is capable of a covalent reaction with component B to form *in situ* the C–X–B copolymer. Reactive compatibilization can only apply to certain selected blend pairs in that at least one blend component must possess a certain functional group capable of reacting with X of the compatibilizer C–X.³

Certain polymers, oligomers, and small molecules containing multiple glycidyl functional groups, C–X_n ($n > 2$), are able to function as effective reactive compatibilizers for certain polymer pairs if both blend constituents A and B contain the necessary functional groups.^{4–10} A fraction of the C–X_n is able to react *in situ* with both A and B simultaneously to form the desired A–X–C–X–B mixed copolymer at the interface and acts as a reactive compatibilizer for the blend of A/B.

From a practical point of view, a readily available compatibilizer without requiring timely consumed synthesis is highly desirable. In this article, we demonstrate another example that a commercially available multifunctional epoxy monomer can effectively compatibilize the immiscible and incompatible blend of poly(ethylene terephthalate) (PET) and poly(phenylene ether) (PPE). Both PET and PPE possess the necessary functional groups (–COOH and phenolic–OH) that are capable of reacting with the multifunctional epoxy monomer.

EXPERIMENTAL

The PETs (IV = 1.0, 0.85, and 0.63) employed were donated by the Shinkong Synthetic Fibers

Correspondence to: F.-C. Chang.
Contract grant sponsor: National Science Council, Republic of China.
© 1997 John Wiley & Sons, Inc. CCC 0021-8995/97/040739-15

Corp. of Taiwan. The unmodified PPE powder was purchased from Generic Electric Co. of the United States which has an intrinsic viscosity of 0.4 dL/g measured in chloroform at 25°C. The tetrafunctional epoxy resin was tetraglycidyl-4,4'-diaminodiphenylmethane (TGDDM), a product from Ciba-Geigy of Switzerland. The difunctional epoxy, DER 332, was obtained from the Dow Chemical Co. of the United States. The catalyst ethyltriphenylphosphonium bromide was purchased from Merck of Germany.

Melt blending was carried out using a 30 mm corotating twin-screw extruder produced by Sino-Alloy Machinery of Taiwan by controlling the barrel temperature between 265 and 290°C. The extruded pellets were dried and injection-molded into standard $\frac{1}{8}$ in. ASTM testing specimens using an Arburg 3-oz injection-molding machine of Germany using temperatures between 290 and 300°C.

The torque vs. time relation was performed in a Brabender Plastic-Corder of Germany at 290°C and 30 rpm. Melt flow rates (MFRs) of the blends were also measured at 290°C using a 2.16 kg load on an instrument from Ray-Ran Co. of England. Capillary rheological properties were carried out using a rheometer from the Kayeness Co. of the United States, Model Galaxy X.

Fourier transform infrared spectroscopic (FTIR) analysis, to detect epoxy groups, was carried out using a Nicolet 500 infrared spectrophotometer (U.S.A.). Thermal properties were investigated by differential scanning calorimetry (DSC) at a

heating rate of 10°C/min immediately after quenching from 300°C (−30°C/min) on a DSC analyzer from Seiko Co. of Japan. The morphologies of the cryogenically fractured surfaces of the injection-molded samples were inspected at the plane perpendicular to the processing flow. The methods to determine the mechanical properties were described previously.^{4,7,9}

RESULTS AND DISCUSSION

Melt Properties

Figure 1 shows the torque versus time curves of the PET and PET/epoxy = 100/1 mixture at 290°C. The high torque observed for PET at 60 s is probably due to the melting of the crystalline PET with a high content of solid PET in the mixture. After about 100 s, the torque values of the PET/epoxy = 100/1 mixture are substantially higher than that of the virgin PET. Polyepoxides have been extensively used as a branching/chain extension of polyesters.^{11,12} The torque increase in the PPE/epoxy system is relatively insignificant as shown in Figure 2, an indication of lower reactivity relative to the previous PET/epoxy system. Figure 3 gives the torque versus time curves of the unmodified and the epoxy-modified PET/PPE = 70/30 blends. The higher torque of the epoxy-modified blend can be attributed to the formation of higher molecular weight species from the reactions between the epoxy with PET and/or PPE.

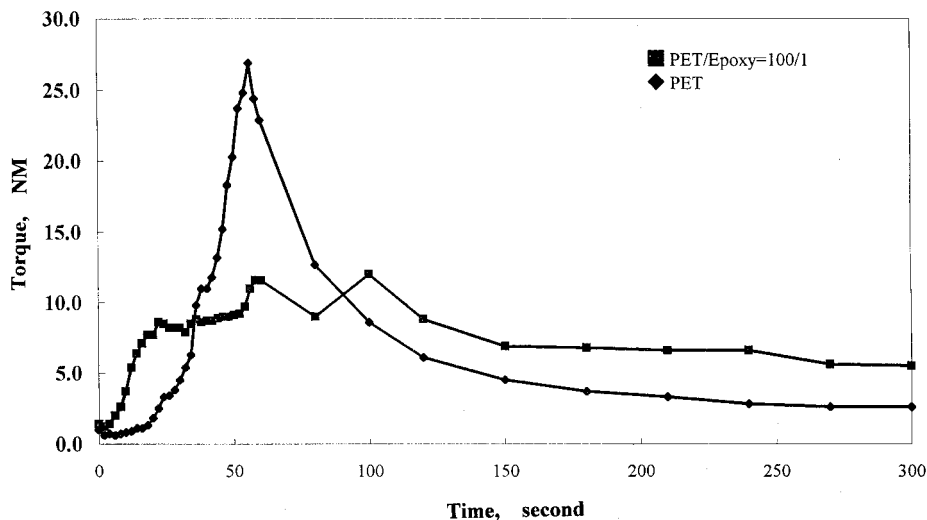


Figure 1 Torque versus time curves of PET and PET/epoxy = 100/1 blend.

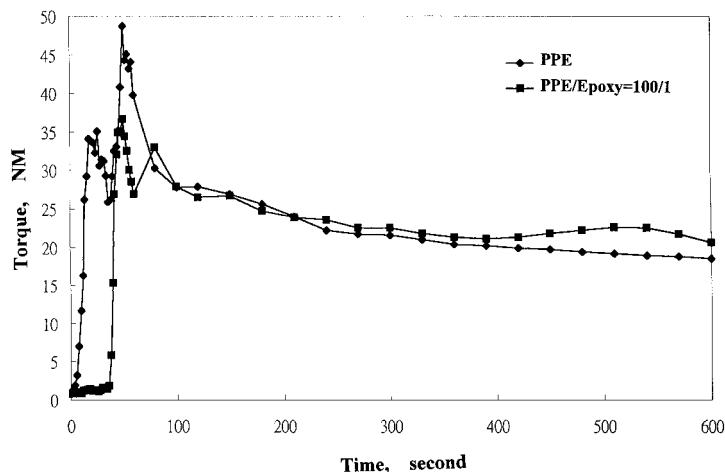


Figure 2 Torque versus time curves of PPE and PPE/epoxy = 100/1 blend.

Figure 4 gives the relation of epoxy content and the corresponding melt flow rate (MFR) of the unmodified and various epoxy-modified PET/PPE = 70/30 blends. The observed MFR decrease with increasing epoxy content comes from the expected reactions mentioned above. A similar trend without any single exception was also been found from other compositions as shown in Table I.

Figure 5 gives the shear rate versus shear viscosity of PET, PET/PPE = 70/30, and the epoxy-modified blends. Again, the capillary rheological results show a higher viscosity from the blend containing a greater quantity of epoxy.

Fourier Transform Infrared (FTIR) Spectroscopy

Figure 6 gives the comparative IR spectra of the PET/epoxy = 100/1 blend before and after melt mixing at 290°C. The disappearance of the epoxy characteristic peak at 910 cm^{-1} can be attributed to the reaction between epoxy and carboxylic acid and hydroxyl end groups of the PET. Figure 7 also shows the disappearance of this epoxy characteristic peak of the PPE/epoxy = 100/1 blend after melt mixing. This epoxy has been demonstrated to be able to react with the phenolic-OH end group of PPE under melt conditions.⁷ This epoxy

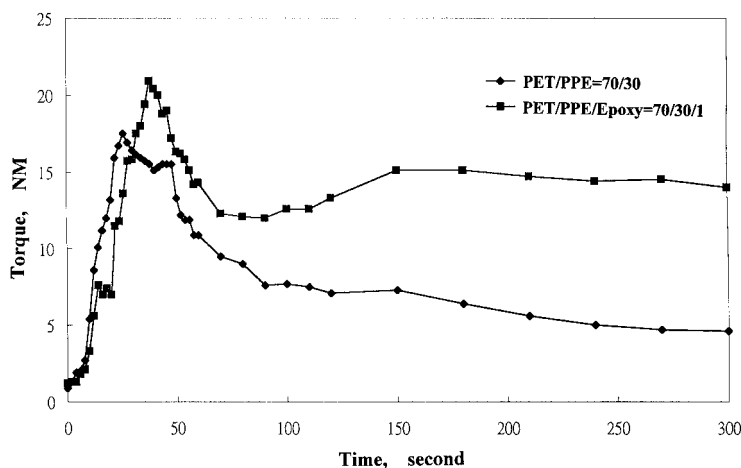


Figure 3 Torque versus time curves of PET/PPE = 70/30 and PET/PPE/epoxy = 70/30/1 blends.

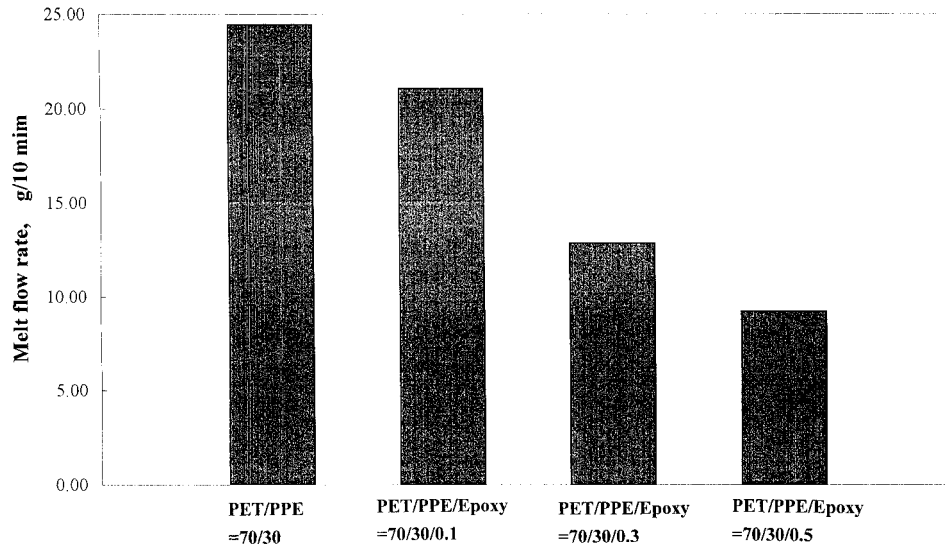


Figure 4 MFR of uncompatibilized and epoxy-compatible PET/PPE = 70/30 blends.

peak also disappeared in the PET/PPE/epoxy = 70/30/1 blend after melting (Fig. 8), as would be expected. Reduction or loss of this epoxy char-

acteristic peak detected by IR spectra does not allow us to identify the expected products from the anticipated reactions between epoxy with

Table I Summarized Data on Melt Flow Rates (MFR) and Mechanical Properties

Composition	MFR (g/10 min)	Unnotch Impact (J/M)	Tensile Strength (MPa)	Tensile Elongation (%)	Flexural Strength (MPa)	Flexural Modulus (MPa)	G_c (J/m ²)
<u>PET (IV = 1)/PPE/epoxy/cat</u>							
90/10	29.5	> 900	53.2	424	87.3	2400	—
90/10/0.3	12.7	> 900	57.3	541	93.3	2480	—
80/20	25.5	> 900	54.6	44	90.6	2460	—
80/20/0.3	17.2	> 900	58.7	84	94.4	2470	—
70/30	24.5	217	55.8	15	89.3	2410	4210
70/30/0.1	21.0	270	56.3	17	92.5	2480	5590
70/30/0.3	12.9	311	57.2	24	95.7	2520	6950
70/30/0.3/0.02	—	374	—	—	—	—	—
70/30/0.5	9.2	281	61.9	11	102.6	2600	9330
50/50	12.4	104	41.0	5.6	80.8	2520	—
50/50/0.3	5.0	195	48.0	6.8	83.4	2510	—
30/70	—	143	58.7	8.7	85.6	2330	—
30/70/0.3	—	244	59.6	8.7	88.9	2340	—
<u>PET (IV = 1)/PPE/DER 332/cat</u>							
70/30/0.3	—	300	—	—	—	—	—
70/30/0.5/0.02	—	324	—	—	—	—	—
<u>PET (IV = 0.85)/PPE/epoxy</u>							
70/30	—	189	52.0	7.6	84.6	2350	—
70/30/0.3	—	429	55.8	9.5	89.3	2350	—
<u>PET (IV = 0.63)/PPE/epoxy</u>							
70/30	—	182	50.8	7.3	85.5	2340	—
70/30/0.3	—	212	52.6	7.6	86.7	2360	—

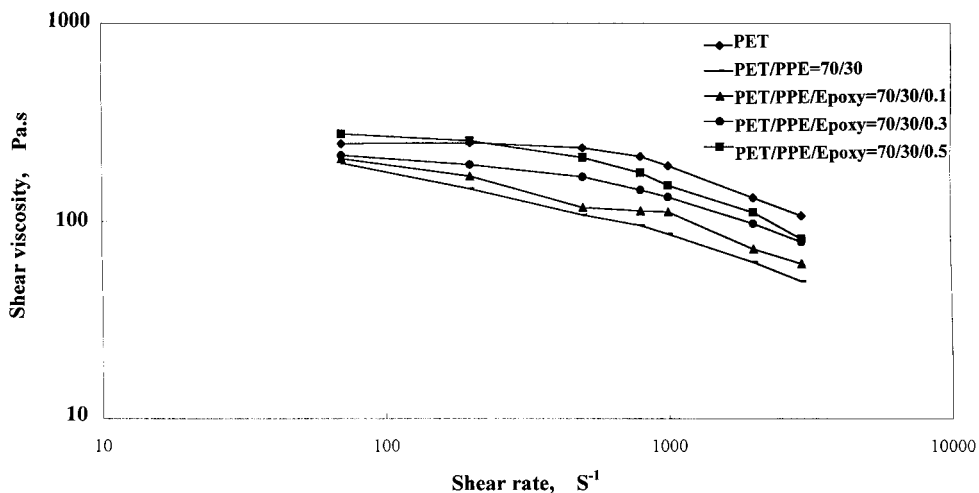


Figure 5 Shear rate versus shear viscosity curves of PET and various epoxy-compatibilized PET/PPE = 70/30 blends.

—COOH and/or —OH end groups because of the instrumental sensitivity limit. Additionally, other reactions such as epoxy hydrolysis could also occur if the system contains even small amount of water.¹³

DSC Analyses

The thermal properties analyzed by DSC for pure components and blends are summarized in Table II. Figure 9 shows the DSC scans of pure compo-

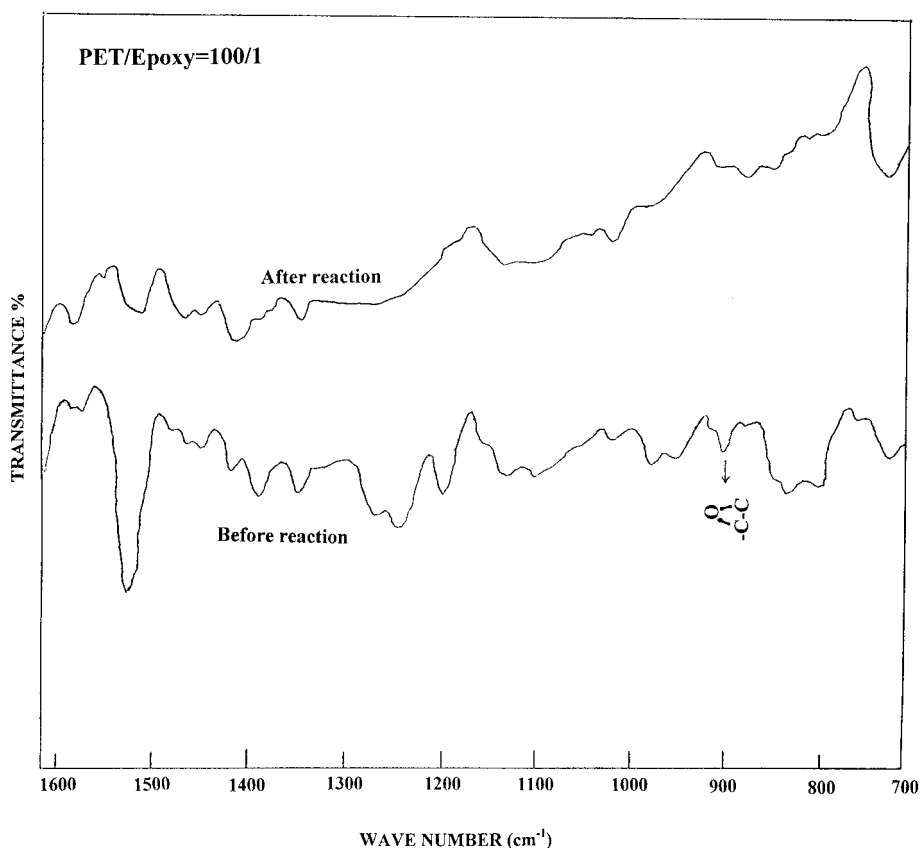


Figure 6 IR spectra of the PET/epoxy = 100/1 blend before and after melt reaction.

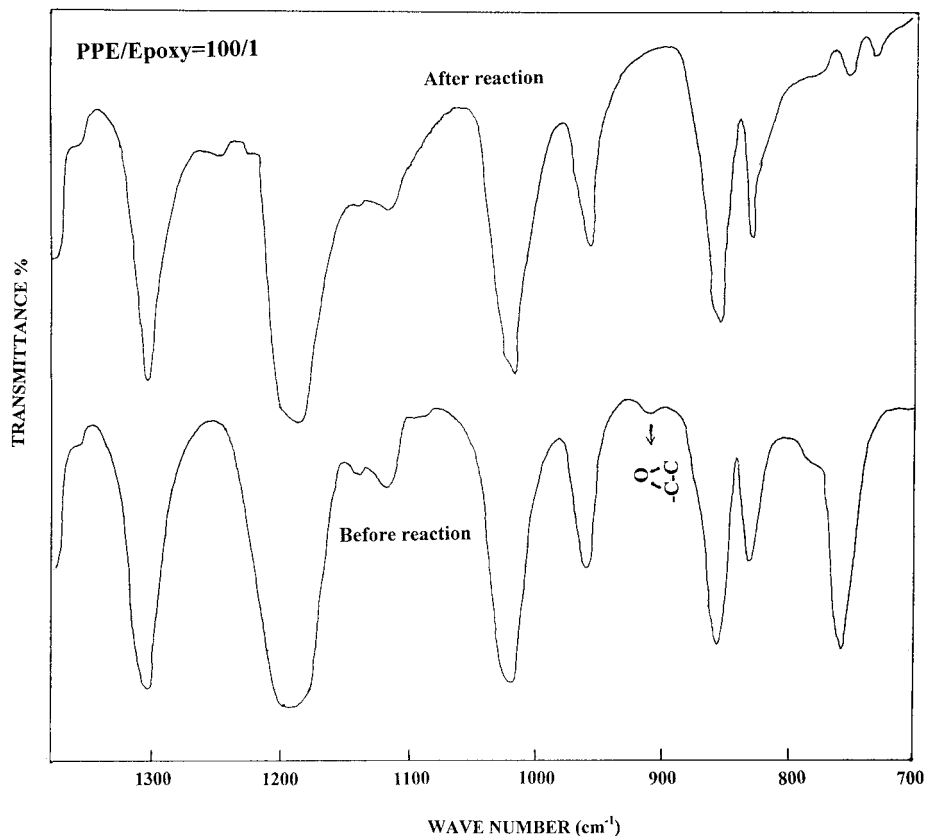


Figure 7 IR spectra of the PPE/epoxy = 100/1 blend before and after melt reaction.

nents and uncompatibilized and compatibilized blends. Glass transition temperatures (T_g s) of PET and PPE tend to shift inward slightly with the addition of the epoxy compatibilizer. Melting temperatures of PET in the blends remain fairly constant regardless of compatibilization. All the uncompatibilized blends have the PET phase already fully crystallized after quenching (at about $-30^\circ\text{C}/\text{min}$ rate) and no crystallization peak can be detected in these DSC heating scans (Fig. 9). The PET crystallization peaks appear on all the compatibilized blends. Figure 10 illustrates the effect of the compatibilizer quantity on thermal properties of the blend based on the PET/PPE = 70/30 blend series. Both crystallization temperature (T_c) and heat of crystallization (ΔH_c) increase with increase of the epoxy content in the blend. Higher T_c during heating scanning of a blend implies a slower crystallization rate because it requires a higher temperature (and longer time) to initiate crystallization. Higher heat release of crystallization (ΔH_c) of a blend also indicates a slower rate of crystallization because a greater fraction of the amorphous PET

left during the process of cooling. Therefore, under an identical thermal treatment condition, a slower crystallization rate always results in a higher T_c and a higher ΔH_c during a DCS heating scanning. Pure PET has the highest T_c and ΔH_c and therefore has the slowest rate of crystallization. The uncompatibilized blends have the highest rate of PET crystallization because no crystallization peak is present. The presence of PPE is able to function as the PET crystallization nuclear agent. On the contrary, a better compatibilized blend tends to reduce its crystallization rate.

SEM Morphologies

Figure 11 gives the SEM micrographs of the uncompatibilized and compatibilized PET (IV = 1)/PPE = 70/30 blends. The dispersed PPE average particle diameter decreases from $4.0\ \mu\text{m}$ for the uncompatibilized blend to 3.0 , 2.3 , and $1.9\ \mu\text{m}$ by the addition of 0.1 , 0.3 , and 0.6 phr epoxy, respectively (Table III). The presence of the $200\ \text{ppm}$ catalyst further decreases the particle size ($1.7\ \mu\text{m}$). Figure 12(A) and (C) shows that the PPE

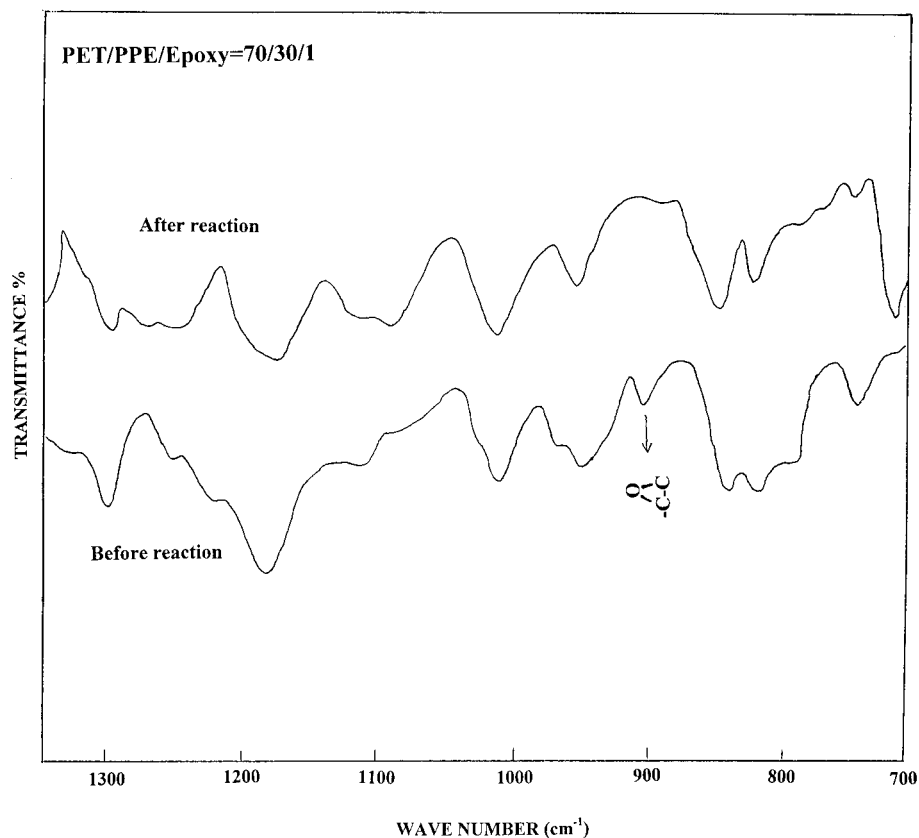


Figure 8 IR spectra of the PET/PPE/epoxy = 70/30/1 blend before and after melt reaction.

Table II Summarized DSC Data

Composition PET (IV = 1)/ PPE/Epox/Cat.	T_{g1} (°C)	T_{g2} (°C)	T_c (°C)	T_m (°C)	ΔH_c (J/g)	ΔH_m (J/g)	Percent PET Crystallinity
100/0	87.4	—	168	261	27.7	32.3	3.8
90/10	87.3	228	—	263	0	26.0	23.6
90/10/0.3	89.8	223	143	262	3.1	27.8	22.5
80/20	91.8	226	—	264	0	26.2	26.8
80/20/0.3	92.3	222	144	263	0.9	26.6	26.4
70//30	89.4	223	—	263	0	19.7	23.0
70/30/0.1	90.7	220	140	264	0.5	21.3	24.3
70/30/0.3	91.4	219	143	263	2.4	22.2	23.2
70/30/0.3/0.02	91.4	218	141	263	0.7	20.1	22.6
70/30/0.5	89.2	216	144	262	3.4	22.7	22.7
50/50	91.6	223	—	263	0	17.4	28.5
50/50/0.3	93.3	222	—	262	3.7	14.9	18.3
30/70	79.7	221	132	262	0	9.3	15.2
30/70/0.3	80.7	222	136	263	3.7	7.0	9.0
0/100	—	224	—	—	—	—	—

T_{g1} , T_g of PET phase; T_{g2} , T_g of PPE phase; T_c , crystallization temperature of PET during heating; T_m , melting temperature of PET; ΔH_c , heat of PET crystallization; ΔH_m , heat of PET melting; percent PET crystallinity is based on the quenched state. X_c (%) = $[(\Delta H_m = \Delta H_c)/\Delta H_{PET}] [100/x]$, where X_c is the PET phase crystallinity in the blend, ΔH_{PET} is the theoretical heat of fusion of the 100% crystallinity of PET; x is the weight fraction of PET in the blend, and $\Delta H_{PET} = 122.25$ J/g (ref. 14).

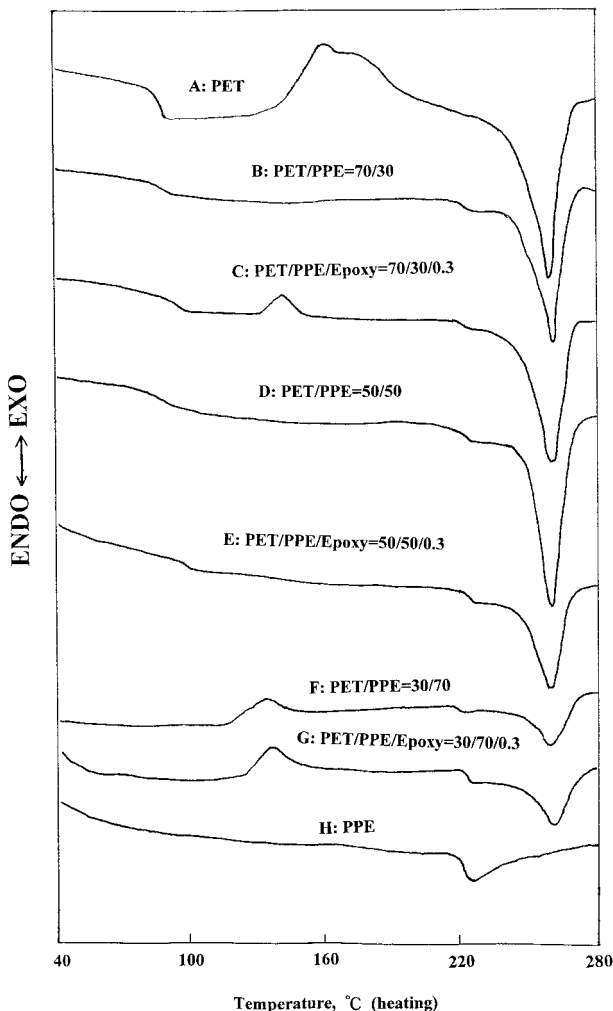


Figure 9 DSC scans of PET, PPE, and the uncompatibilized and compatibilized PET/PPE blends.

particle size increases with decrease of the PET IV value for those uncompatibilized blends. The viscosity ratio of the dispersed drop vs. the matrix ($P = \eta_d/\eta_m$) near unit gives the smallest dispersed particles.¹⁵ The viscosity of PPE is approximately six times higher than that of PET (IV = 1) and a lower PET IV would result in an even higher viscosity mismatch and therefore a greater dispersed particle size. The measured average particle diameters from the uncompatibilized and compatibilized PET/PPE = 70/30 blends by varying the IV of PET are listed in Table III.

Wu¹⁵ previously proposed a relationship between the critical Weber number ($G\eta_m a/\gamma$) and the viscosity ratio (P) as follows:

$$G\eta_m a/\gamma = 4(\eta_d/\eta_m)^{0.84} \quad (\text{for } P > 1) \quad (1)$$

where G is the shear rate; a , the dispersed particle

diameter; and γ , the interfacial tension. To predict the dispersed particle diameter (a) to compare with the experimentally obtained particle size, component viscosities (η_d and η_m), shear rate (G), and interfacial tension (γ) are required. The shear rate can be calculated from the geometry of the processing equipment and processing conditions (such as the extruder rpm), but the interfacial tension is relatively difficult to obtain. However, the relative particle size from the blends with various viscosity ratios (by changing matrix viscosity) can be obtained if the processing conditions are identical. Equation (1) for blends 1 and 2 can be rearranged into

$$a_1/a_2 = (\eta_{m2}/\eta_{m1})^{0.84} \quad (2)$$

where a_1 and a_2 are the diameters of the dispersed particles for blends 1 and 2. The viscosity of pure PPE is not necessary in determining the relative particle size as indicated in eq. (2). The shear rate in a corotating twin-screw extruder at a screw speed of 200 rpm was estimated in the order

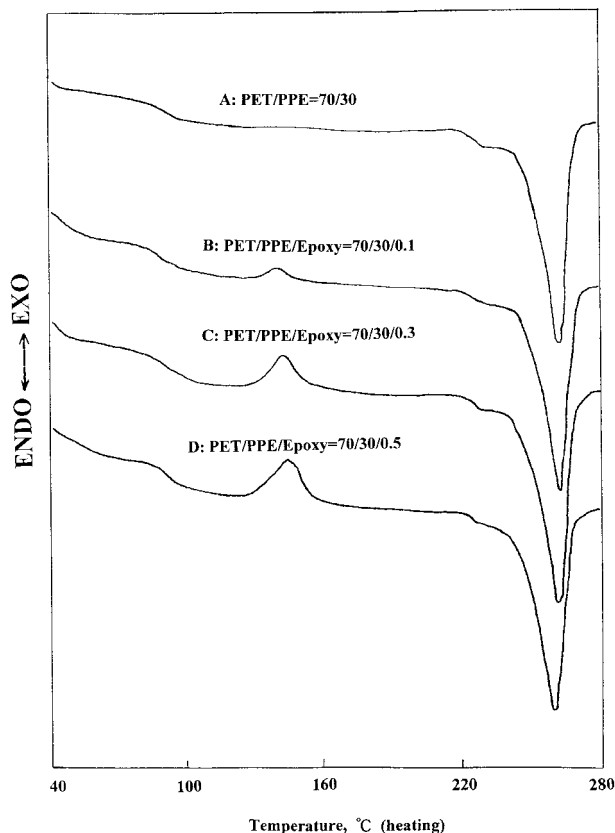
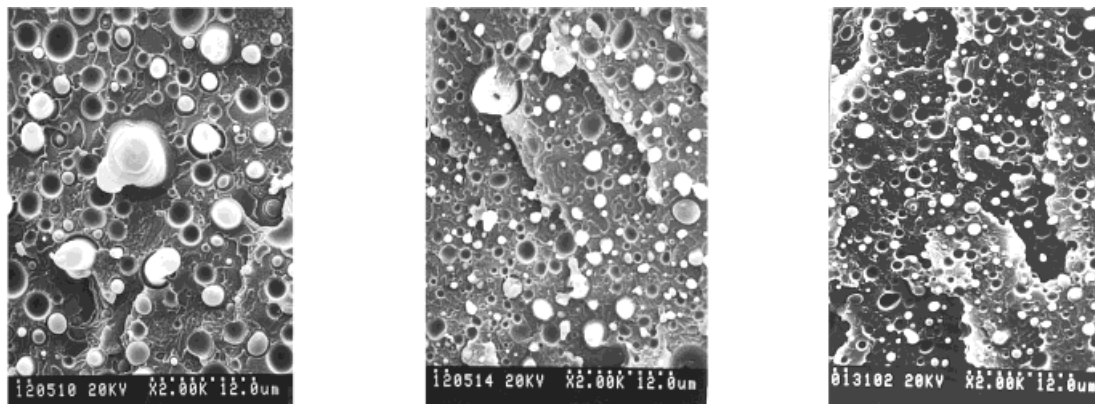


Figure 10 DSC scans showing the effect of epoxy content on PET/PPE = 70/30 blends.



A: PET(IV=1)/PPE=70/30

B: PET(IV=1)/PPE/Epoxy=70/30/0.3

C: PET(IV=1)/PPE/Epoxy/Cat.=70/30/0.3/0.02

Figure 11 SEM micrographs of the uncompatibilized and compatibilized PET (IV = 1)/PPE = 70/30 blends: (A) PET (IV = 1)/PPE = 70/30; (B) PET (IV = 1)/PPE/epoxy = 70/30/0.3; (C) PET (IV = 1)/PPE/epoxy/Cat = 70/30/0.3/0.02.

of 200 s^{-1} .¹⁵ Shear viscosities of PETs with various IV were determined by a capillary rheometer at 200 s^{-1} shear rate and the results are listed in Table III. For those uncompatibilized PET/PPE = 70/30 systems, the calculated particle diameter ratios are fairly close to those from experiment as shown in Table III.

Interfacial tension is one the major factors in dictating the dispersed particle size in the blend. Ideally, the interfacial tension between the blend constituents PET and PPE can be calculated from their individual surface tension provided that they are known.¹⁶ Alternatively, the interfacial tension can be calculated from the measured interfacial thickness between two phases.¹⁶

$$\gamma = 7.6L^{-0.88} \quad (3)$$

where L is the interfacial thickness. The above two methods require knowledge of either the component surface tensions or the interfacial thickness, which are beyond the scope of this study. If

we choose the PET (IV = 1)/PPE = 70/30 series containing different amounts of epoxy (and catalyst) by assuming a constant viscosity ratio (η_d/η_m), we can estimate relative interfacial tension from the measured particle diameter ratio by the equation⁴

$$\gamma_2 = \gamma_1(a_2/a_1) \quad (4)$$

Figure 13 shows the plot of relative interfacial tension vs. epoxy content in the uncompatibilized and compatibilized PET/PPE = 70/30 blends. The presence of a catalyst further decreases the interfacial tension of the blend. Figure 14 shows the SEM micrographs on the PET/PPE = 30/70 and 50/50 series where the domain size has been reduced after compatibilization.

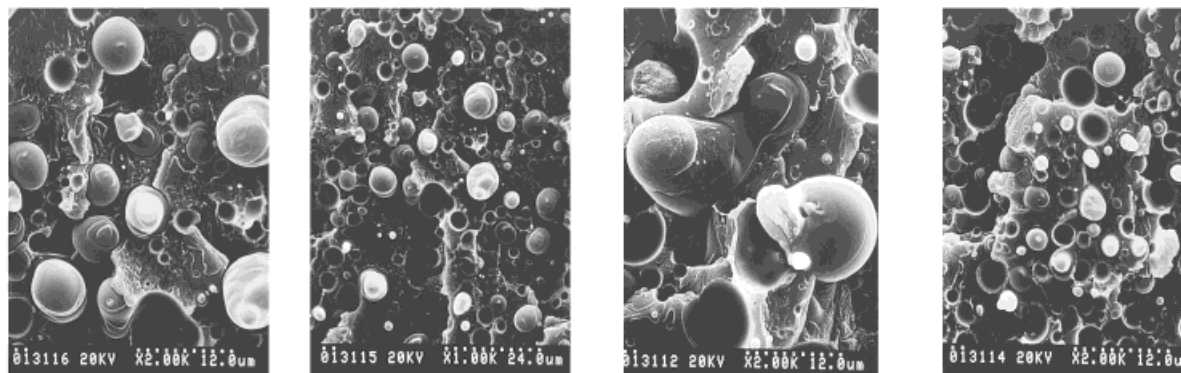
Mechanical Properties

Mechanical properties including the unnotch impact, tensile, flexural, and strain energy release

Table III Particle Diameters and Calculated Relative Particle Diameter Ratios

Blend Series	IV of PET	η_{PET} (Pa-s)	Particle Diameter of PET/PPE/Epoxy/Cat (μm)				
			70/30	70/30/0.1	70/30/0.3	70/30/0.5	70/30/0.3/0.02
1	1.0	270	4	3	2.3	1.9	1.7
2	0.85	150	7	—	4.2	—	—
3	0.63	100	10	—	5.4	—	—

η_{PET} measured at 290°C and shear rate of 200 s^{-1} . Relative particle diameter ratio: $a_1/a_2 = 0.57 \mu\text{m}$ (exp), $0.61 \mu\text{m}$ (cal); $a_1/a_3 = 0.40 \mu\text{m}$ (exp), $0.43 \mu\text{m}$ (cal).



A: PET(IV=0.85)/PPE=70/30 B: PET(IV=0.85)/PPE/Epoxy=70/30/0.3 C: PET(IV=0.63)/PPE=70/30 D: PET(IV=0.63)/PPE/Epoxy=70/30/0.3

Figure 12 SEM micrographs of the PET/PPE = 70/30 series containing lower PET intrinsic viscosity: (A) PET (IV = 0.85)/PPE = 70/30; (B) PET (IV = 0.85)/PPE/epoxy = 70/30/0.3; (C) PET (IV = 0.63)/PPE = 70/30; (D) PET (IV = 0.63)/PPE/epoxy = 70/30/0.3.

rate (G_c) are summarized in Table I. Only the PET/PPE = 70/30 series will be discussed and the results should be able to apply to other compositions. Unnotch impact is more appropriate to use to differentiate the toughness change of the notch sensitive blends through compatibilization. Figure 15 shows the effect of epoxy content and the presence of catalyst on the unnotch impact strength. The impact strength increases with increasing epoxy content and reaches a maximum at 0.3 phr; further increase of epoxy content results in lower impact strength. The presence of 200 ppm of the catalyst results in further improvement in impact strength. A similar trend has also been obtained on other compositions as shown in Table I. The difunctional epoxy DER 332 also im-

proves the impact properties but is less effective than is the tetrafunctional epoxy TGDDM (Table I). In general, the blend containing lower IV of the PET (IV = 0.85 or 0.63) results in lower impact strength, as would be expected.

Figure 16 shows that the tensile strength increases with increasing epoxy content. For example, the tensile strength of the uncompatibilized PET/PPE = 70/30 blend increases from 55.8 to 61.9 MPa after the addition of 0.5 phr of the epoxy compatibilizer. The corresponding blends containing lower IV of PET results in a relatively lower tensile strength as shown in Table I. Tensile elongation shows a similar trend as to that the impact strength; the maximum elongation occurs at 0.3 phr epoxy content (24%, Table I or Fig. 17).

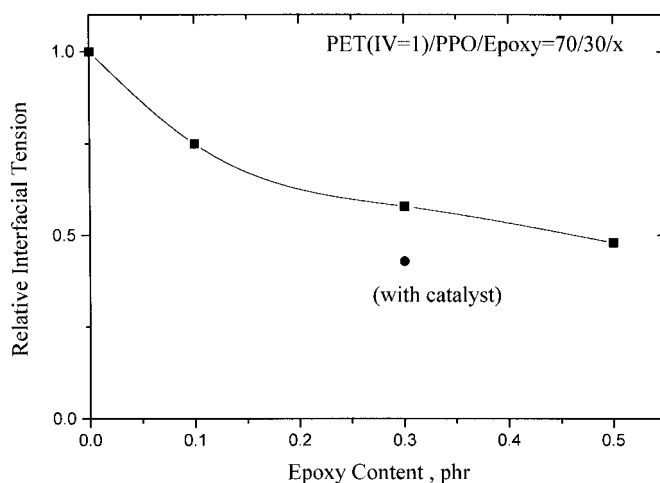


Figure 13 Plot of relative interfacial tension versus epoxy content on the PET/PPE = 70/30 blends.

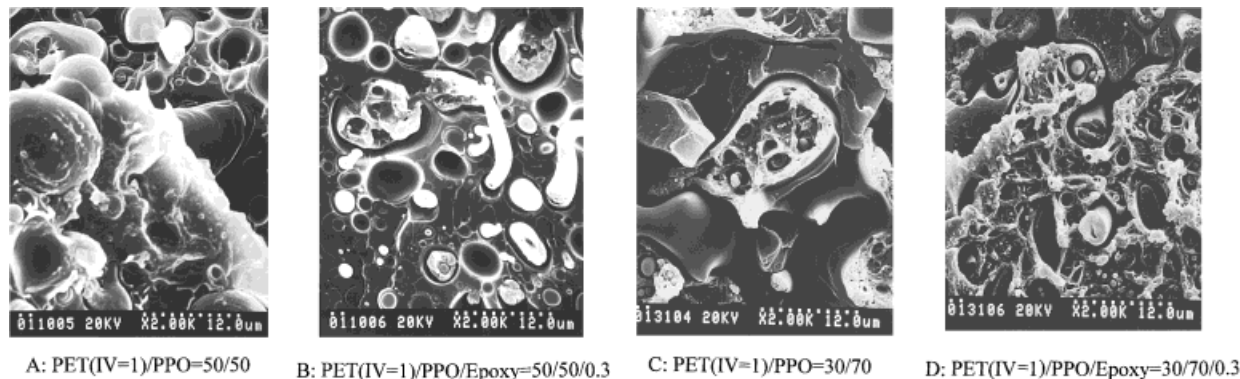


Figure 14 SEM micrographs of the uncompatibilized and compatibilized PET/PPE = 50/50 and PET/PPE = 30/70 blends: (A) PET (IV = 1)/PPE = 50/50; (B) PET (IV = 1)/PPE/epoxy = 50/50/0.3; (C) PET (IV = 1)/PPE = 30/70; (D) PET (IV = 1)/PPE/epoxy = 30/70/0.3.

Tensile elongations from those blends containing lower IV of the PET component are substantially lower (Fig. 18).

The trend of flexural strength is very similar to that of the tensile strength as shown in Table I. The flexural modulus also increases with increase of the epoxy content (Table I).

Figure 19 shows that the critical strain energy release rate (G_c) increases with increase of the epoxy content. The G_c of the uncompatibilized PET/PPE = 70/30 blend increases from 4210 to

9330 J/m² by the addition of 0.5 phr of the epoxy compatibilizer.

Overall, the mechanical property improvements through this epoxy compatibilizer are very substantial when we consider that only a very small amount of the compatibilizer was employed relative to the conventional compatibilizer. In comparing with a similar system using the conventional reactive compatibilizer styrene-glycidyl methacrylate copolymer to compatibilize PBT/PPE blends, this low molecular weight epoxy

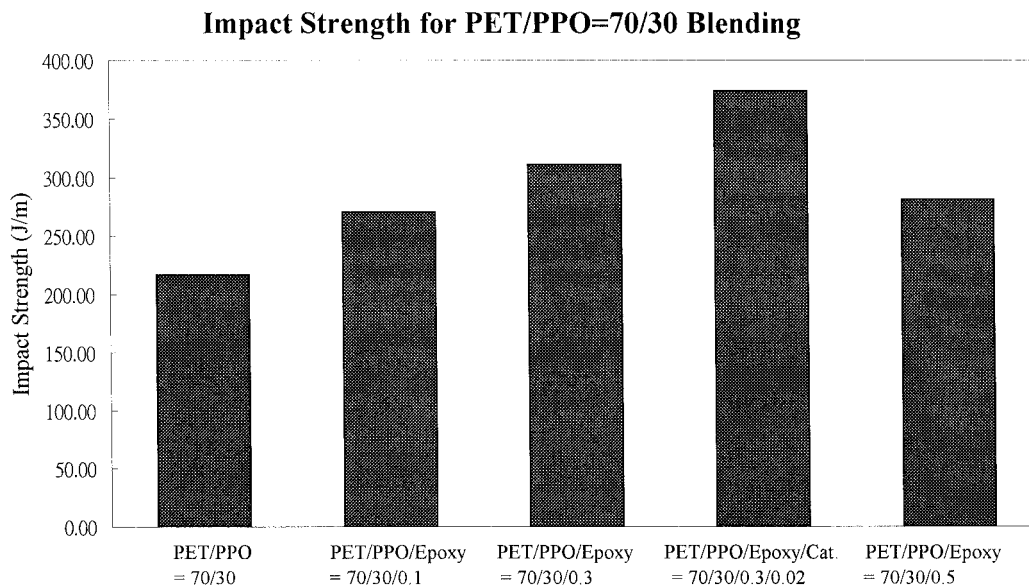


Figure 15 Effect of epoxy content and catalyst on the unnotch impact strength of the PET/PPE = 70/30 blends.

Tensile Strength PET/PPO Blending System

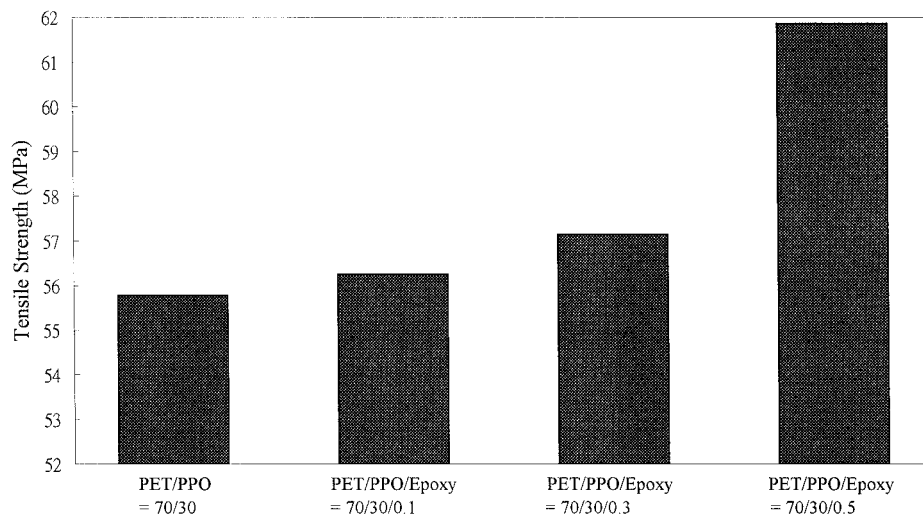


Figure 16 Effect of epoxy content on the tensile strength of the PET/PPE = 70/30 blends.

needs only about 10–20% by weight relative to the conventional system to achieve the same level of compatibilization.

Compatibilizing Mechanism

The mechanism of a nonreactive compatibilization system is relatively simple; it involves the migration of the nonreactive block or graft copoly-

mer into the interface. The conventional reactive compatibilization is more complex and the related mechanism was discussed in detail elsewhere.³ The coupler-type compatibilization is the most complicated system, since it involves reactions with both matrix components simultaneously. Since a reactive compatibilizer itself structurally does not function as a surfactant of the blend, it is considered as a precursor of the true compatibi-

Elongation at Break for PET/PPO=70/30 Blending System

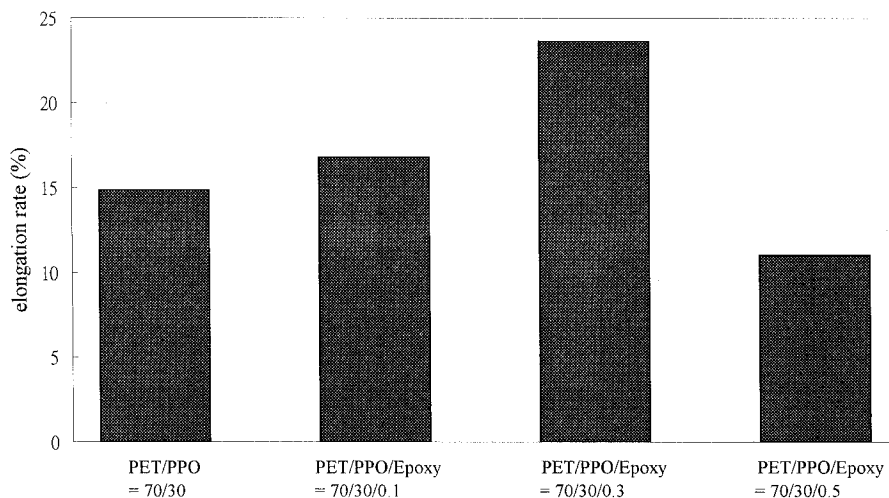


Figure 17 Effect of epoxy content on the tensile elongation of the PET/PPE = 70/30 blends.

Elongation at Break for PET/PPO=70/30 Blending System

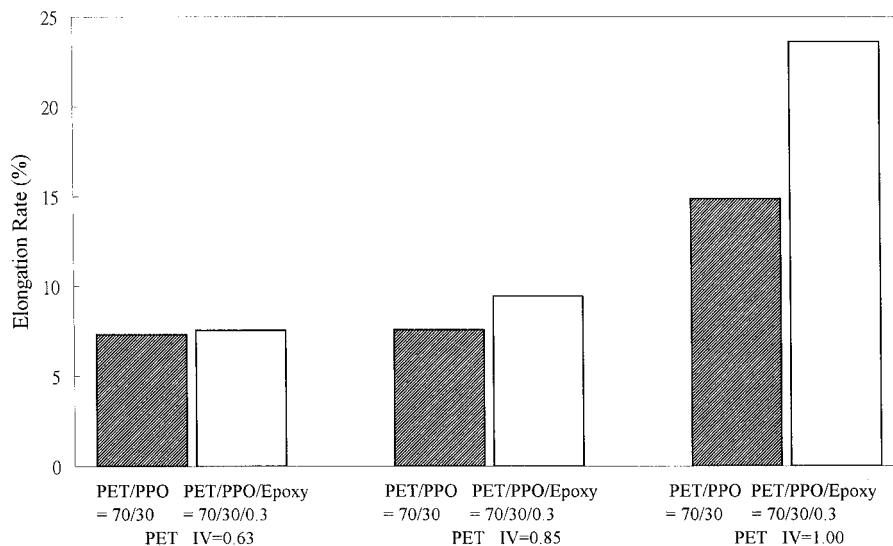


Figure 18 Effect of the PET intrinsic viscosity on tensile elongation of the PET/PPE = 70/30 blends.

lizer. Only the desirable product, a mixed copolymer containing segments from both blend components, can be considered as an effective compatibilizer. In a couple-type compatibilization system, many factors such as type of reaction, extent of reaction, when the reaction occurs, and the final

location of the reacted copolymer are complicated but important in designing an optimized compatibilization system. Therefore, several variables have to be taken into consideration including compatibilizer functionality and quantity, relative reactivity and compatibility with both blend constit-

Critical Strain Energy Release rate (PET/PPO=70/30)

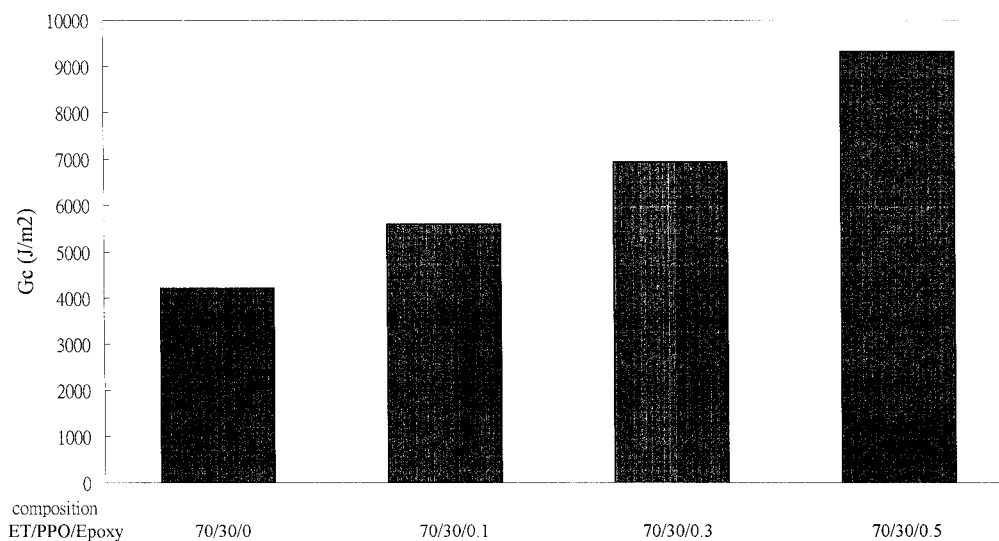


Figure 19 Effect of epoxy content on the critical strain energy release rate (G_c) of the PET/PPE = 70/30 blends.

uents, processing condition, blending sequence, glass transition, and melting temperatures of the blend components. The most ideal situation of an effective coupler-type compatibilization system can be described as follows:

1. The compatibilizer has lower compatibility with both blend components and prefers to reside at interface.
2. Each compatibilizer molecule reacts with both components simultaneously and lightly.
3. The resultant mixed copolymer anchors along the interface.

In reality, only a fraction of the initially added compatibilizer is able to form the desirable mixed copolymer and anchors at the interface. In a worst situation, the compatibilizer may reside and consume completely in one particular phase (more compatible and higher reactivity one); the resulting products tend to reside in that phase and it may lose its role as a phase compatibilizer. In this case, the epoxy simply functions as chain extender or branching agent.

This tetrafunctional epoxy TGDDM is more efficient in compatibilizing the PET/PPE blends than is the difunctional DER 332 based on a better mechanical property improvement and smaller phase domains. Statistically, a molecule containing four functional groups has a better opportunity to react with both blend matrices to form the desired mixed copolymer than that from two molecules, each containing two functional groups.

The relative compatibility between the reactive compatibilizer with blend components usually can be estimated by comparing their individual solubility parameters. In this system, the solubility parameters of PET, PPE, and epoxy are 10.9, 9.0, and 7.3–14.6 Hildebrand, respectively, which are too close to differentiate their relative compatibilities. Therefore, we assume that the epoxy is partially miscible with PET and PPE with comparable compatibility.

The melting temperature of PET (261°C, Table I) is higher than the T_g of PPE (224°C, Table I). Therefore, epoxy tends to dissolve into the PPE phase in the earlier stages of extruder blending and has the first opportunity to make contact and react with PPE. Reactivity between epoxy with PPE is relatively lower than that with PET based on the previous torque vs. time data. This may be advantageous; otherwise, most or all the epoxy functional groups might have been con-

sumed with PPE if the reactivity between epoxy with PPE were significantly higher than that with PET.

Since the relative compatibilities between epoxy with PET and with PPE are assumed to be comparable, these epoxy molecules containing four functional groups have a good chance to make contact and react with both PET and PPE simultaneously to form the desirable PET–epoxy–PPE mixed copolymer. Once this mixed copolymer is formed, it tends to anchor at the interface because it contains both PET and PPE segments.

A certain fraction of the epoxy molecules may react only with one particular blend component, PET or PPE, and the resultant product tends to reside in that respective phase. In such a situation, the reacted product cannot function as a compatibilizer. It serves as a chain extender or branching agent. The formation of the PET–epoxy–PPE mixed copolymer is believed responsible for the high efficient compatibilization achieved by this relatively lower molecular weight tetrafunctional epoxy.

CONCLUSIONS

This commercially available tetrafunctional epoxy has been demonstrated as an highly efficient reactive compatibilizer for PET/PPE blends. It requires a very small quantity (about 10–20%) of the epoxy compatibilizer relative to most conventional compatibilizers to gain the same level of compatibilization. This epoxy has the opportunity to make contact and react with both PET and PPE simultaneously during high shear extruder melt mixing to form the desirable PET–epoxy–PPE mixed copolymer. This mixed copolymer contains both PET and PPE segments that tend to anchor along the interface to function as an effective compatibilizer.

This work was financially supported by the National Science Council, Republic of China.

REFERENCES

1. M. Xanthos and S. S. Dagli, *Polym. Eng. Sci.*, **31**, 929 (1991).
2. N. C. Liu and W. E. Baker, *Adv. Polym. Technol.*, **11**, 249 (1992).

3. F. C. Chang, in *Handbook of Thermoplastics*, O. Olabisi, Ed., Marcel Dekker, New York, 1996, Chap. 21, pp. 491–521.
4. C. C. Huang and F. C. Chang, *Polymer*, to appear.
5. H. C. Chin, K. C. Chiou, and F. C. Chang, *J. Appl. Polym. Sci.*, **60**, 2503 (1996).
6. H. C. Chin and F. C. Chang, *Polymer*, to appear.
7. C. R. Chiang and F. C. Chang, *J. Appl. Polym. Sci.*, **61**, 2411 (1996).
8. D. Y. Chang and F. C. Chang, *J. Appl. Polym. Sci.*, **56**, 1015 (1995).
9. W. B. Liu, W. F. Kuo, C. R. Chiang, and F. C. Chang, *Eur. Polym. J.*, **32**, 91 (1996).
10. J. An, J. Ge, and Y. Liu, *J. Appl. Polym. Sci.*, **60**, 1803 (1996).
11. W. F. H. Borman and J. A. Rock, U.S. Pat. 4,020,122 (1977) (to General Electric).
12. Y. Arai and T. Tanaka, U.S. Pat. 4,520,175 (1985) (to Unitika).
13. C. T. Maa and F. C. Chang, *J. Appl. Polym. Sci.*, **49**, 913 (1993).
14. D. W. Van Krevelen, *Properties of Polymers*, Elsevier, Amsterdam, 1976.
15. S. Wu, *Polym. Eng. Sci.*, **27**, 335 (1987).
16. S. Wu, *Polymer Interface and Adhesion*, Marcel Dekker, New York, 1982.