

Use of Epoxy Reactivity for Compatibilization of PP/PBT and PP/LCP Blends

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SYNOPSIS

Binary blends of a reactive ethylene-based terpolymer with polybutylene terephthalate (PBT) and with a liquid crystalline polyester (LCP) were studied to clarify the possible interactions between the blended polymers. The aim was to determine the suitability of the reactive terpolymer containing epoxy reactivity as a compatibilizer in blends of polypropylene (PP) and these two polyesters. The binary blends exhibited increased viscosity during blending, changes in the crystallization of the PBT phase, and an intimate contact between the blended polymers, which pointed to strong interactions or chemical reactions between the compatibilizer and both PBT and LCP. FTIR analysis confirmed the reaction of the epoxide and formation of new esters. Most probably the carboxyl end groups of the polyesters reacted with the epoxy group of the compatibilizer. In the second part of the work the same terpolymer was shown to act as a compatibilizer in PP/PBT and PP/LCP blends. This behavior was based on good mixing with the PP phase and on the chemical reactivity or strong interactions with the polyesters demonstrated in the investigations on binary blends. Addition of 5 wt % of the compatibilizer improved the impact strength, especially in PP/PBT blends where synergistic behavior was found at compositions of 80/20 and 20/80. In PP/LCP blends, the compatibilizer significantly improved the impact strength of unnotched samples at 20 wt % LCP content. In both blends, the compatibilizer reduced the size of the dispersed domains and caused them to attach better in the matrix.

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INTRODUCTION

Blending two or more engineering thermoplastics is a useful way to obtain new materials with desired property combinations. Although most engineering polymers are immiscible and even incompatible with each other, useful blends with good properties can be achieved through formation of special morphologies. Effective compatibilization is required to bind the discrete blend components together and to achieve toughness.

Reactive extrusion can be used for compatibilizing dissimilar polymers. Functional end groups or specially functionalized polymers react with corresponding reactive groups in the other polymer, and new covalent bonds are formed during the extrusion

process. Often, even strong physical interactions or hydrogen bonding may be enough to achieve sufficient interfacial adhesion between the dispersed polymer and the matrix.

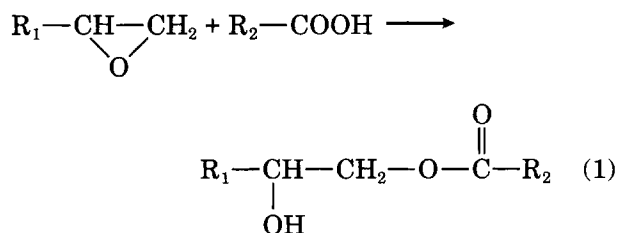
Blending of polyolefins with engineering polymers such as thermoplastic polyesters is a useful way to upgrade the properties of polyolefins, and can offer both price and property advantages. One major problem that limits the direct blending of polyolefins with engineering polymers is the nonpolar structure of polyolefins. However, reactor-made olefin-based co- and terpolymers with functional groups are commercially available, and many functionalities can be introduced into the structure of polyolefins through postmodification. Such functionalized polyolefins may act as compatibilizers between polyolefins and high-performance engineering polymers.

Reactions between carboxyl acid and epoxy groups have been exploited in many blends of thermoplastic polyesters.¹ Epoxy-modified poly-

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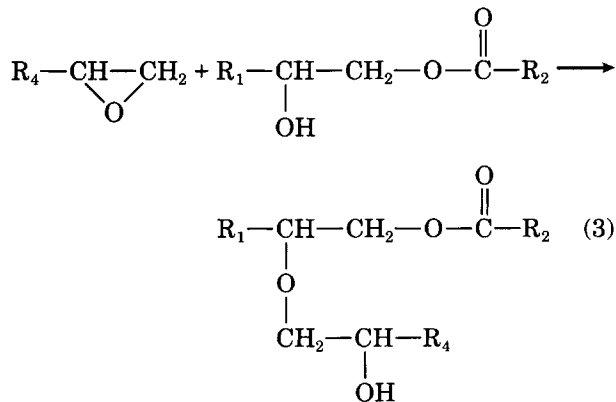
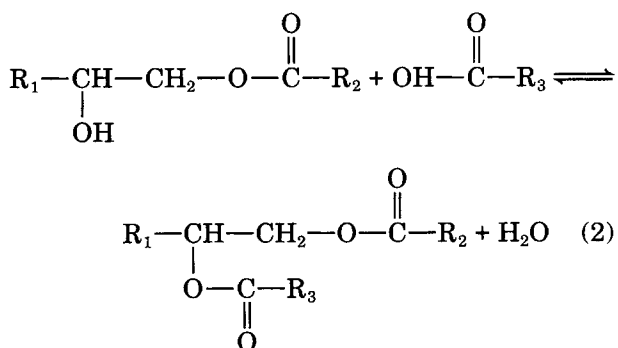
styrene (PS) has been used for compatibilization of blends of polyphenylene ether (PPE) and polybutylene terephthalate (PBT).² Glycidyl methacrylate grafted polyphenylene ether (PPE-*g*-GMA) reacts with acid-terminated polybutylene terephthalate.³ Impact modification of thermoplastic polyesters can be achieved through blending with epoxy-containing elastomers. In particular, glycidyl methacrylate grafted EPDM is often used for this application.⁴ The addition of an epoxy-containing terpolymer consisting of ethylene, acrylic ester, and glycidyl methacrylate has been found to significantly improve the impact properties of polybutylene terephthalate and polyethylene terephthalate (PET).⁵

The strong polarization of the O—H bond of carboxylic acids ensures fast reaction between epoxy and carboxylic acid groups.



The epoxy-carboxyl reaction leads to the formation of hydroxyl side groups, which can be very useful for further chemical modification of polymer chains such as grafting or crosslinking. It is an addition reaction and no volatile compounds are formed.

In principle, at least three other reactions are possible; namely, esterification (2) or etherification (3) of secondary hydroxyl groups and hydrolysis of the epoxide group. The probability of these reactions occurring depends on temperature, catalyst, and the nature of the reaction medium. Their contribution is generally higher when the reaction is carried out in bulk than when carried out in solution.^{6,7}



Polybutylene terephthalate (PBT) is characterized by good thermal and mechanical properties, excellent surface appearance, and stable electrical properties. One major drawback is its poor notched impact strength. For blending of polybutylene terephthalate, compatibilizers with electrophilic functionalities capable of reacting with the nucleophilic end groups of polyesters are recommended. Compatibilizers with acrylic acid, maleic anhydride, or epoxy groups are often mentioned as potential candidates for modifying the properties of thermoplastic polyesters through blending with polyolefins.^{8,9} Also, styrene/ethylene-butylene/styrene triblock copolymer can be used for compatibilization of polyethylene terephthalate/polyethylene and other such blends.¹⁰

Thermotropic liquid crystalline polymers (LCPs) have received a lot of attention during recent years as a new class of high-performance polymers. When small amounts of LCPs are blended with thermoplastics, *in situ* composites may be formed, with improved material properties such as enhanced strength and stiffness, thermal resistance, dimensional stability, and processability.¹¹⁻¹³ Blending of LCPs, in particular with low-cost polyolefins, would be of both technical and commercial interest. Besides their good properties, LCP blends generally have one major negative feature: poor impact strength, which is mainly due to the lack of interfacial adhesion between the blend components. Suitable compatibilization is, thus, needed to overcome the brittleness and to achieve useful blends. Only a few compatibilization studies on blends of LCPs and polyolefins have so far been reported.^{14,15}

Although epoxy groups are frequently mentioned as potential candidates for reactions with end groups of polyesters, few systematic attempts have been made to specify compatibilization effects and specific interactions in blends containing polyesters as one

component. The aim of this work was to investigate the effects of an epoxy functionalized compatibilizer on the morphology and properties of PP/LCP and PP/PBT blends. The interactions between the compatibilizer and the two blend components were of particular interest. In the first part of the work, these interactions were studied by analyzing binary blends of the compatibilizer with PBT and with LCP. In the second part, the compatibilizer was added to PP/PBT and PP/LCP blends to discover its effects on the final blend properties and morphology.

EXPERIMENTAL

Materials

The epoxy functionalized polymer, ethylene/ethyl acrylate/glycidyl methacrylate (E/EA/GMA) terpolymer Lotader AX 8660, was supplied by Atochem. The ethyl acrylate and glycidyl methacrylate components together account for 32% of the total composition.⁵ The melting temperature is 79°C, and the melt index is 6.0 g/10 min (190°C/2.16 kg). This polymer is later referred to as the compatibilizer. Polybutylene terephthalate (PBT) was Grilpet XE 3060 produced by EMS Chemie, with a melting temperature of 219°C and density of 1.32 g/cm³. The thermotropic liquid crystalline polymer (LCP) was Vectra A 950 produced by Hoechst Celanese. This is a totally aromatic polyester-type LCP based on 6-hydroxy-2-naphthoic acid (HNA) and *p*-hydroxy benzoic acid (HBA). It has a melting temperature of 280°C and density of 1.40 g/cm³. Polypropylene (PP) VB 1950K was supplied by Neste and has a melt index of 1.9 g/10 min (2.16 kg, 230°C).

Blending and Processing

Binary Blends

Binary interactions between the compatibilizer and PBT and between the compatibilizer and LCP were investigated in samples prepared in a Brabender W 50 EH batch mixer. The compositions studied were 100/0, 70/30, 50/50, 30/70, and 0/100. Before blending, the polyesters were predried in a Conair Micro-D dehumidifying dryer, PBT 5 h at 120°C and LCP over night at 100°C. All blends were made under nitrogen atmosphere. For PBT/compatibilizer blends, the temperature was 240°C or 270°C and the rotation speed 50 or 80 rpm. For LCP/compatibilizer blends, a temperature of 300°C and ro-

tating speed of 50 rpm were applied. Mixing time was kept constant at 15 min. During mixing, torque was measured as a function of time. Pure polymers were treated in a similar manner to the reference.

Compatibilized Blends

Compatibilized PP/PBT and PP/LCP blends were investigated with corresponding noncompatibilized compositions as references. The compositions (by weight) of the binary blends were 100/0, 80/20, 60/40, 40/60, 20/80, and 0/100. The corresponding compatibilized blends were made by adding 5 wt % of the ethylene-based compatibilizer containing glycidyl methacrylate.

The blends were made with a corotating twin-screw extruder Berstorff ZE 25 × 33D. The melt temperature was 240°C for PP/PBT blends and 290°C for PP/LCP blends. After blending, the materials were dried over night at 90°C before injection molding to test specimens with an Engel ES 200/40 injection molding machine.

Characterization

The differential scanning calorimetry (DSC) measurements were made on the binary blends by using Mettler DSC-30 equipment with a heating and cooling rate of 10°C/min.

The morphology of the fractured surfaces of all the blends was investigated with a JEOL JXA-840A scanning electron microscope (SEM). The samples were fractured after dipping in liquid nitrogen and the fractured surfaces were coated with a 15 nm layer of gold.

Fourier transform infrared spectroscopy (FTIR) was applied to characterize the changes in the chemical structure of the polymers after blending. Very thin samples were cut with a microtome, and the FTIR analysis was made with an IR microscope. A fine resolution of 0.5 cm⁻¹ was used to detect even slight shifts in peaks.

Samples for rheological measurements were made with a counterrotating twin-screw extruder (Brabender DSK 42/7) at the same temperatures as used in the batch mixer and with a screw speed of 50 rpm. Melt viscosities in the shear flow were measured with a Göttfert Rheograph 2002 capillary viscosimeter at 240°C for PBT/compatibilizer and at 290°C for LCP/compatibilizer blends. The L/D ratio of the die was 20/1 mm. The results include Rabinowitch correction, but Bagley correction was not made. The melt viscosities of the compatibilized and noncompatibilized blends were measured in a similar way.

Flexural properties of the injection molded blends were measured according to ISO 178 by the three-point bending test with a crosshead speed of 5 mm/min. Notched Charpy impact strength was determined according to ISO 179. For unnotched samples, an applied Charpy test was used where the dimensions of test bars were $4 \times 10 \times 112$ mm and the span was 70 mm. It should be noted that these dimensions slightly differed from those of the standard test (ISO 179).

RESULTS AND DISCUSSION

Interactions in Binary Blends

Torque Measurements during Mixing

The torque was recorded during blending in a Brabender batch mixer as a means of detecting changes of melt viscosity. Lamba¹⁶ and Xanthos¹⁷ interpret the increase in viscosity during mixing as an indication of reactions. The torques of binary blends of PBT and the compatibilizer are illustrated in Figure 1. At all compositions studied the blends exhibited higher torques than the pure reference polymers. In the blends containing 30 wt % compatibilizer in PBT the torque level began to increase significantly after 4 min of mixing. The same kind of torque behavior was observed in blends of LCP and the compatibilizer; a similar increase in torque occurred at 30 wt % compatibilizer but now reaching the level of 20 Nm.

The high level of torque and, thus, viscosity of the binary blends is probably due to a reaction between the epoxy groups in the compatibilizer and carboxylic end groups of the polyesters. This reaction creates free OH groups, which, again, are reactive with epoxy groups. During a long time of mixing the described reaction may also lead to considerable branching and finally networking of the epoxy-containing terpolymer through crosslinks initiated by carboxyl terminated groups in PBT or LCP. These last reactions do not necessarily improve the compatibility between the blended polymers. At the composition 70/30 there are more COOH end groups available for the reaction with the compatibilizer than at the two other compositions. This is probably the reason for the remarkable increase of torque at this particular composition. Effects of temperature and rotating speed on the torque are presented in Figure 2.

The reactions proceeded at a higher rate when mixing was more vigorous. Higher temperature, in turn, led to lower torques and did not promote the reactions. Perhaps the crosslinking reactions could not be observed due to thermal degradation. The probability of unwanted side reactions in the reactive extrusion process is, nevertheless, low because the residence time in the twin-screw extruders used for blending is only about 1.5–2 min.

Melt Viscosities

Melt viscosities of the preblended binary blends gave partly the same kind of information about the re-

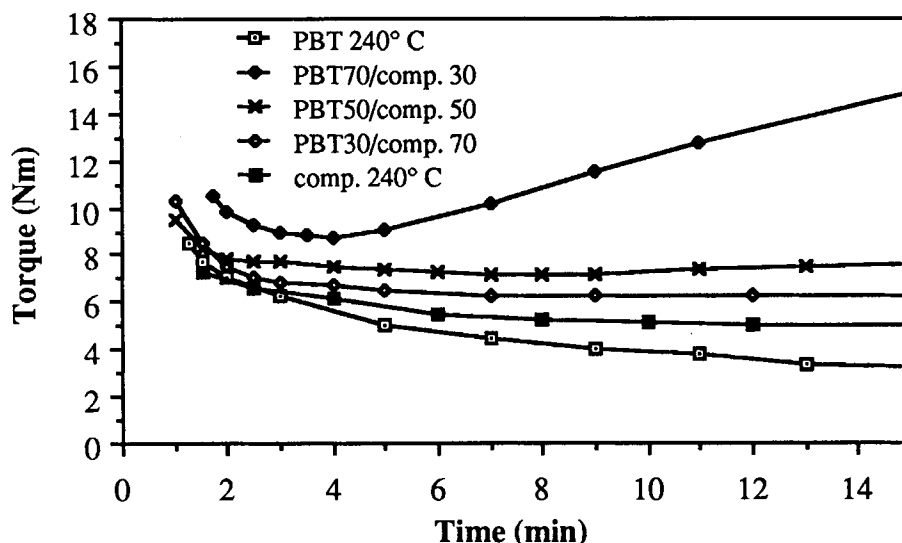


Figure 1 Torque of PBT, the compatibilizer, and their blends during mixing in a Brabender batch mixer at 240°C and with rotating speed of 50 rpm.

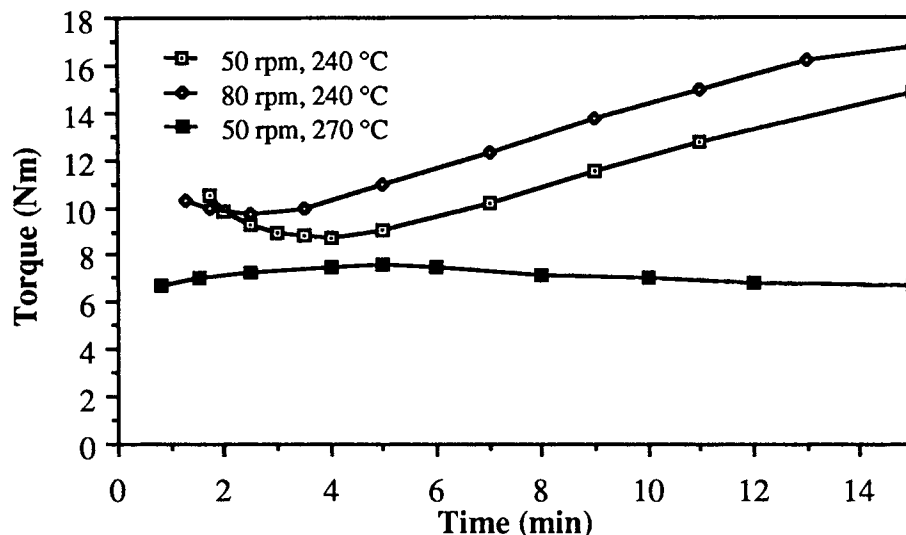


Figure 2 Effects of temperature and rotating speed on the torque of a PBT/compatibilizer 70/30 blend mixed in a Brabender batch mixer.

actions and possible crosslinking as did the continuous torque measurement during the mixing stage. The shear rates applied were much higher, however, than those in the batch mixer and, hence, corresponded better to the shearing conditions of the actual processing. On the other hand, the long measuring times in the capillary rheometer exceeded the residence times in extrusion or injection molding. Melt viscosities of PBT, the compatibilizer, and their binary blends are shown in Figure 3(a). Again, the blend containing 30 wt % of the compatibilizer exhibited a high viscosity, reaching the level of pure PBT, even though the pure compatibilizer had a very low viscosity. Also, at other compositions the blends had higher viscosities than expected from the rule of mixtures.

An even higher viscosity increase was found for the blends of LCP and the compatibilizer, shown in Figure 3(b) at a composition 80/20. With higher contents of the compatibilizer than 20 wt %, the melt viscosities of blends of LCP and compatibilizer were too high to be measured. Again, the increase was related to the chemical reactions during the blending. The long measuring time may also contribute to crosslinking reactions.

The high viscosities of the binary blends of LCP/compatibilizer and PBT/compatibilizer were indicative of chemical reactions between the compatibilizer and the two polyesters. As noted above, besides the addition reaction of the compatibilizer and carboxylic acid end groups [eq. (1)], crosslinking may occur and, through increased molecular weight, contribute to the higher viscosity. The compatibil-

izer had no effect on the viscosity of the nonreactive polypropylene.

DSC Results

DSC analysis of binary blends did not indicate significant changes in the melting behavior of the individual polymers. The most important information obtained from the DSC curves was contained in the crystallization thermograms of PBT/compatibilizer blends shown in Figure 4. The crystallization temperature of pure PBT was 190°C, but in the blend containing 30 wt % of the compatibilizer the crystallization peak was located at about 184°C, and in the blend with 50 wt % of the compatibilizer at 152°C. In the latter blend, there was also a smaller crystallization peak at 100°C. In the blend with 70 wt % of the compatibilizer, only the crystallization peak at 100°C was observed. The kind of fractionated crystallization of the dispersed phase evident in Figure 4 has earlier been shown to correlate with enhanced compatibility and diminished size of the dispersed particles.^{18,19} Thus, the changes in crystallization behavior of PBT can be interpreted as further evidence of interactions between the two polymers. In the LCP/compatibilizer blends no changes in crystallization nor in the glass transition temperature of LCP could be observed because of difficulties in determining these things from the DSC data.

Morphology

Morphologies of the binary blends of PBT and the compatibilizer blended in a Brabender batch mixer

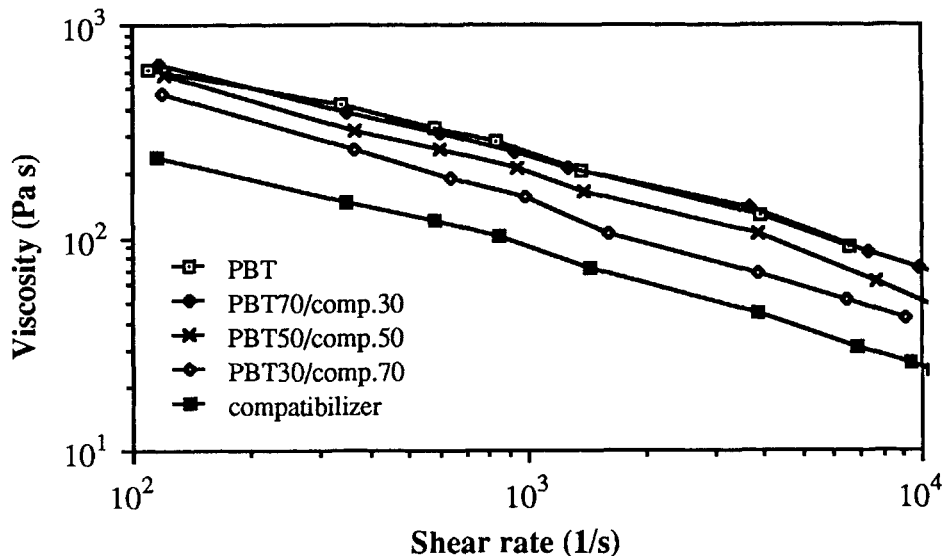


Fig. 3 a

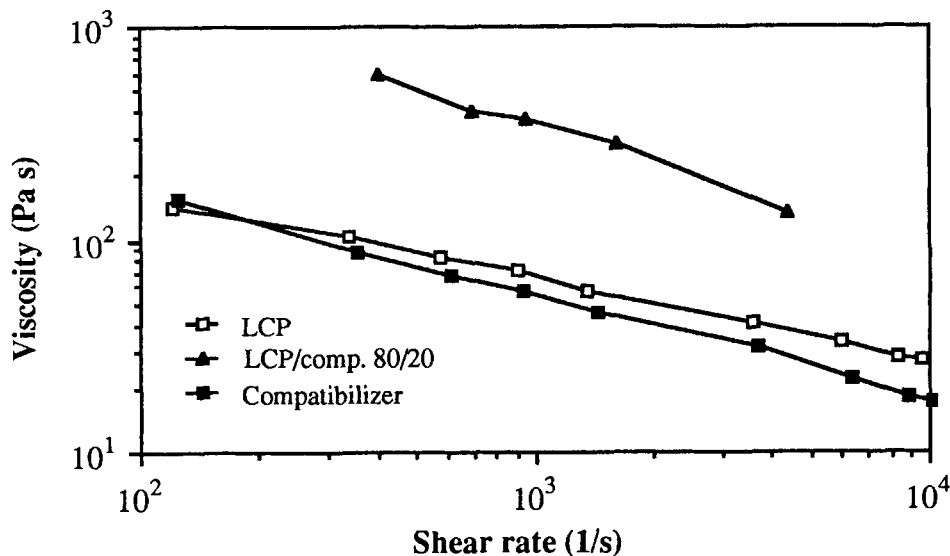


Fig. 3 b

Figure 3 Melt viscosities of (a) PBT, compatibilizer, and PBT/compatibilizer binary blends at 240°C vs. shear rate and of (b) LCP, compatibilizer, and their 80/20 blend vs. shear rate at 290°C.

confirmed the existence of strong interactions between the two polymers. SEM micrographs showed a very fine dispersion in the PBT/compatibilizer 50/50 blend where the diameter of the dispersed compatibilizer particles was less than 1 μm [Fig. 5(a)]. The dispersion was less visible in the LCP/compatibilizer 50/50 blend, and the dispersed particles were very well embedded in the matrix [Fig.

5(b)]. This was an indication of good compatibility and probable reactions between the two polymers. In the second part of the work we studied compatibilized blends where polypropylene was a major component. The morphology of a binary PP/compatibilizer 95/5 blend is, therefore, shown in Figure 5 as well. The ethylene-based terpolymer formed a fine dispersion in the polypropylene matrix, which

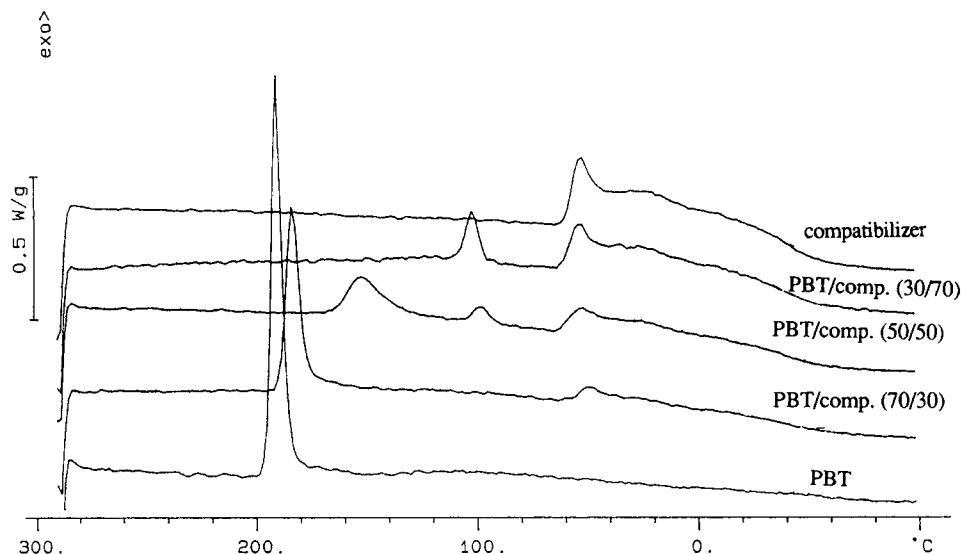


Figure 4 Crystallization thermograms of PBT, compatibilizer, and PBT/compatibilizer binary blends recorded at a sweep rate of $10^{\circ}\text{C}/\text{min}$.

indicates, as expected, that this compatibilizer mixes well with polypropylene. The average particle size of the dispersed compatibilizer droplets was well below $0.5\ \mu\text{m}$.

FTIR Results

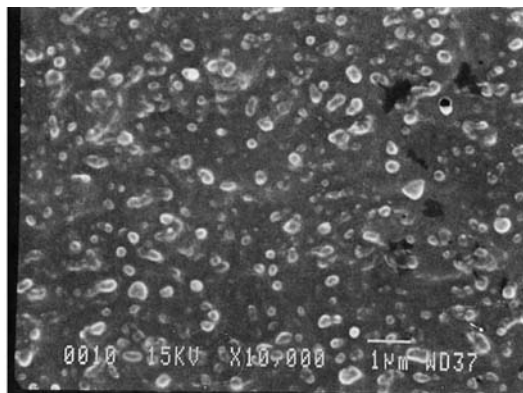
FTIR spectroscopy has been utilized by many research groups^{20,21} to characterize strong interactions such as hydrogen bonding in polymer blends. In this work, the technique was applied to obtain information about chemical reactions that might have taken place during the mixing process. FTIR analysis was applied to the binary blends mixed for 15 min in a Brabender batch mixer. The spectra of the blends were compared with those of pure PBT, LCP, and compatibilizer.

The interpretation of the FTIR curves was not straightforward, because the compatibilizer gave rise to only a few peaks and the interesting peaks of the epoxy groups were difficult to observe not only in blends but in the pure compatibilizer. Primary attention was, consequently, paid to peaks related to ester groups. The difference spectrum technique was used to reveal even small shifts of peaks. A further problem was the high absorptivity of PBT that dominated in blends, and changes in the compatibilizer phase were sometimes hidden behind the PBT trace.

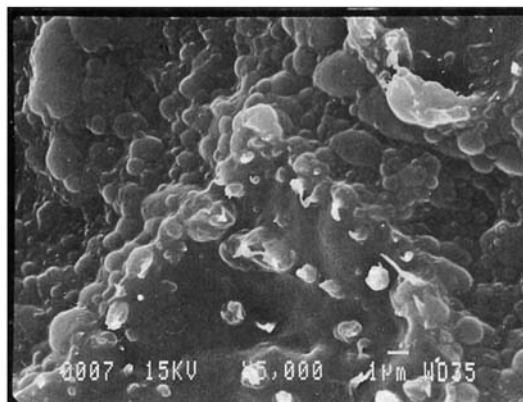
The spectra Nos. 1–3 in Figure 6(a) are difference spectra of PBT/compatibilizer blends minus PBT. If no chemical changes or interactions had occurred, these spectra should have been identical with the spectrum of the compatibilizer. Figure 6(a) shows, however, changes in both the location and intensity of the peaks. Because PBT did not produce peaks in this area, the changes must have been due to chemical changes in the compatibilizer. The changes in the peaks were related to stretching of the C—O—C bonds in the compatibilizer, probably due to reactions of ester groups or the formation of new esters. In addition, there were some shifts related to the deformation vibrations of the aliphatic CH_2 bonds; these shifts might be due to changes in the crystallization of the PBT phase induced by the compatibilizer.

Blends of LCP and the compatibilizer exhibited clearer changes relative to the pure polymers than did PBT blends. Figure 6(b) shows a difference spectrum (No. 2) of the 50/50 blend minus LCP, which should be the same as that of the compatibilizer (No. 1). The clear shifts and changes around $1200\text{--}1100\ \text{cm}^{-1}$ found for the presented peaks and related to asymmetric stretching of the C—O—C bonds point to either chemical reactions or very strong physical interactions such as hydrogen bonding. This finding is supported by another small shift in the peak

(a)



(b)



(c)

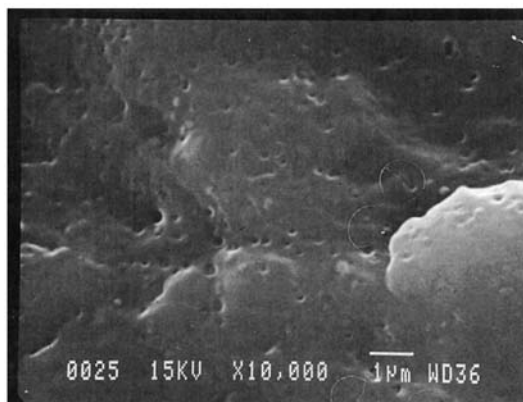


Figure 5 SEM micrographs of binary blends of (a) PBT/compatibilizer and (b) LCP/compatibilizer at a composition of 50/50, and (c) PP/compatibilizer at a composition of 95/5.

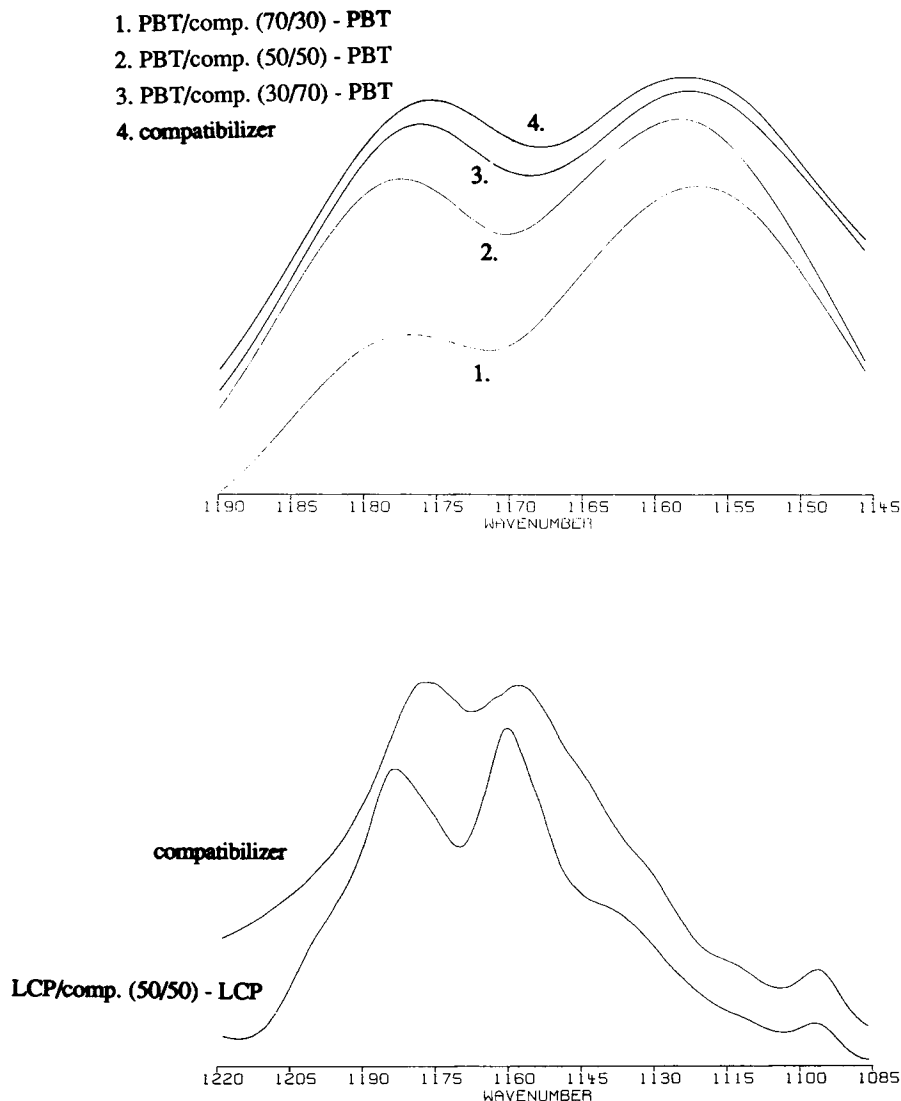


Figure 6 Parts of the FTIR spectra of binary blends: (a) PBT/compatibilizer and (b) LCP/compatibilizer.

around 1090 cm^{-1} related to symmetric stretching of the same bonds.

FTIR analysis of the binary blends confirmed that the changes in morphology and increase in viscosity of the binary blends were due to chemical reactions between compatibilizer and the polyesters.

Compatibilized Blends

Mechanical Properties

Table I summarizes the results of the flexural and impact tests carried out on compatibilized and noncompatibilized PP/PBT blends. The increase in flexural strength and modulus with increased PBT content followed fairly well the linear rule of mix-

tures. Addition of the compatibilizer improved the impact properties as expected, but the level of flexural strength and modulus was slightly lowered at all blend compositions. This was due to the softness and perhaps too large amount of compatibilizer.

Because the noncompatibilized reference samples showed good impact strength in tests with unnotched samples, the notched samples were tested as well. The improvement in toughness evident through compatibilization was even more pronounced in the notched Charpy impact strength (Fig. 7). This was especially seen at compositions far from the phase inversion area, i.e., 80/20 and 20/80.

The presence of the minor phase as a small dispersion at these blend compositions seems to play

an important role in the toughening. These blends exhibited synergistic behavior in notched impact strength exceeding the values of both pure polymers with 5 wt % of the compatibilizer. Similar observations have been reported for polypropylene/polyamide 6 blends with maleic anhydride grafted styrenic block copolymer as the compatibilizer.²² The bimodal behavior in blend toughness can be explained through an analogy with rubber toughening. A fine enough dispersion of the minor component in the matrix allows the formation of localized stress concentrations, which absorb the impact energy through yielding.

The mechanical properties of PP/LCP blends are set out in Table II. As can be seen, the increase of flexural modulus followed the linear rule of mixtures, but the flexural strength was significantly lower than the calculated average values in both the compatibilized and noncompatibilized blends. The level of both strength and modulus was higher than in PP/PBT blends, but did not reach the values achieved in our earlier studies.¹¹⁻¹³ Note that the injection molding conditions were not optimized for each blend composition, but similar conditions were applied to all blends to avoid differences due to the processing step. This might explain the lower than expected strengths at higher LCP contents. The fibrillation and orientation of the dispersed LCP phase can be maximized in extrusion, allowing significant reinforcing.¹²

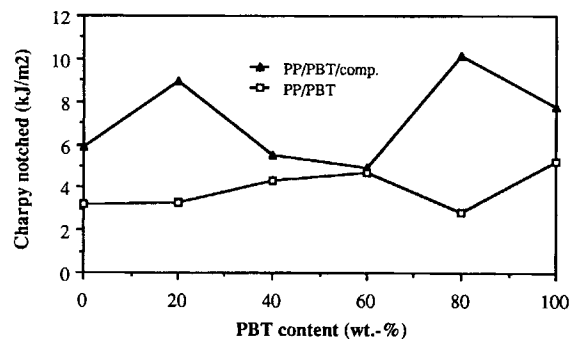


Figure 7 Charpy notched impact strength of PP/PBT blends with and without compatibilization.

In spite of enhanced strength and modulus, the noncompatibilized LCP blends are generally brittle in impact. This is partly due to the anisotropic morphology but mainly to the lack of interfacial adhesion.

At a low concentration (20 wt %) of LCP, the compatibilizer addition improved the impact strength of the unnotched samples as hoped. Similar improvement was not, however, observed in tests with the notched samples. The difference between the PP/PBT and PP/LCP blends in this property may be associated with the totally different morphologies of the two blend systems. The notched Charpy impact strength of PP/LCP blends increased at higher LCP content, as shown in Figure

Table I Flexural and Impact Properties of Injection-Molded PP/PBT Blends

Material	E (MPa)	σ (MPa)	Charpy Impact Strength (kJ/m ²)	
			Unnotched ^a	Notched ^b
PP	929 (40)	28.8 (0.7)	NB	3.2 (0.2)
PP/PBT	80/20	1296 (23)	NB	3.3 (0.2)
PP/PBT	60/40	1386 (21)	46.4*	4.3 (0.4)
PP/PBT	40/60	1555 (17)	50.0*	4.7 (0.6)
PP/PBT	20/80	1748 (14)	85.4*	2.8 (0.1)
PBT	1938 (9)	63.4 (0.2)	NB	5.2 (0.5)
PP/comp	95/5	871 (12)	NB	5.9 (1.6)
PP/PBT/comp	80/20/5	1046 (31)	NB	8.9 (1.2)
PP/PBT/comp	60/40/5	1227 (12)	NB	5.5 (0.5)
PP/PBT/comp	40/60/5	1253 (15)	NB	4.9 (0.1)
PP/PBT/comp	20/80/5	1558 (30)	NB	10.1 (1.0)
PBT/comp	95/5	1858 (20)	NB	7.8 (0.5)

^a Dimensions of the unnotched test bar: 4 × 10 × 112 mm; span: 70 mm; hammer 4 J.

^b Hammer 1 J.

* Only some samples broken.

NB = not broken.

Standard deviations in parentheses.

8. This was related to the enhanced fibrillation of the LCP phase at higher concentrations.

Effects of Compatibilization on the Morphology

Effects of compatibilizer addition on the blend morphology were investigated by scanning electron micrographs of the twin-screw extruded strands. Addition of an appropriate compatibilizer usually decreases the average size of the dispersed particles. This was very evident in the PP/PBT blends. Figure 9 shows the SEM micrographs of PP/PBT 20/80 and 80/20 blends with and without compatibilization. When PBT was the minor phase [Fig. 9(a) and (b)] the compatibilizer did not diminish the particle size to any great extent but made the PBT domains attach extremely well to the PP matrix. On the other hand, when PP was the dispersed phase [Fig. 9(c) and (d)], the compatibilizer reduced the average particle size from about 1–2 μm to 0.3–0.5 μm . In addition, the PP particles seemed to adhere better to the matrix in the compatibilized blend.

The morphology of the PP/LCP blends was even more dramatically changed through the compatibilization. All LCP blends exhibited a skin/core morphology with thin fibers in the skin region and more spherical LCP domains in the core. The effects of compatibilization were better seen in the core. The morphologies of the 20/80 and 80/20 PP/LCP blends (core) with and without 5 wt % of the compatibilizer are shown in Figure 10. At the lower LCP content (20 wt %), the compatibilizer addition re-

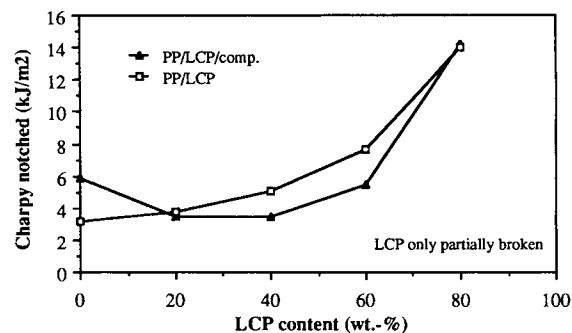


Figure 8 Charpy notched impact strength of PP/LCP blends with and without compatibilization.

duced the particle size from about 2–3 μm to about 0.5 μm [Fig. 10 (a) and (b)]. In addition, the LCP domains were better attached to the matrix in the compatibilized blend. The improved impact strength discussed above was related to these effects. On the other hand, the fine dispersion of LCP in the matrix made the fiber formation more difficult, which can be seen as less fibers in the skin layer and lower than expected strength and stiffness.

At the higher LCP content [Fig. 10 (c) and (d)], the LCP phase was well fibrillated and the dispersed PP was difficult to detect. Owing to the highly fibrillar morphology the samples could not be broken transversally to the fiber direction. Hence, the longitudinally fractured surfaces were analyzed. The highly fibrillar morphology seen in the micrographs [Fig. 10 (c) and (d)] was responsible for the good

Table II Flexural and Impact Properties of Injection-Molded PP/LCP Blends

Material	E (MPa)	σ (MPa)	Charpy Impact Strength (kJ/m ²)	
			Unnotched ^a	Notched ^b
PP	823 (23)	26.2 (0.2)	NB	3.2 (0.2)
PP/LCP 80/20	1881 (40)	41.9 (0.3)	24.6	3.8 (0.3)
PP/LCP 60/40	2809 (83)	47.0 (0.6)	13.0P	5.1 (1.1)
PP/LCP 40/60	4033 (670)	53.7 (3.8)	14.4P	7.7 (0.1)
PP/LCP 20/80	5751 (81)	68.5 (1.1)	21.0P	14.0 (0.8) ^c
LCP	6843 (152)	148.7 (1.4)	85.2P	79.1 (6.5) ^{aP}
PP/comp 95/5	791 (19)	25.0 (0.3)	NB	5.9 (1.6)
PP/LCP/comp 80/20/5	1015 (37)	29.3 (0.7)	NB	3.5 (0.9)
PP/LCP/comp 60/40/5	2582 (178)	47.3 (2.7)	12.6	3.5 (0.2)
PP/LCP/comp 40/60/5	3750 (277)	52.6 (5.6)	15.3P	5.5 (0.3)
PP/LCP/comp 20/80/5	4904 (121)	66.9 (3.6)	21.3P	14.2 (3.5) ^c
LCP/comp 95/5	5539 (151)	120.7 (1.2)	88.9P	71.5 (5.2) ^{aP}

Dimensions of the unnotched test bar: 4 × 10 × 112 mm; span: 70 mm.

Hammer ^a 4 J, ^b 0.5 J, ^c 1 J.

P = partially broken.

NB = not broken.

Standard deviations in parenthesis.

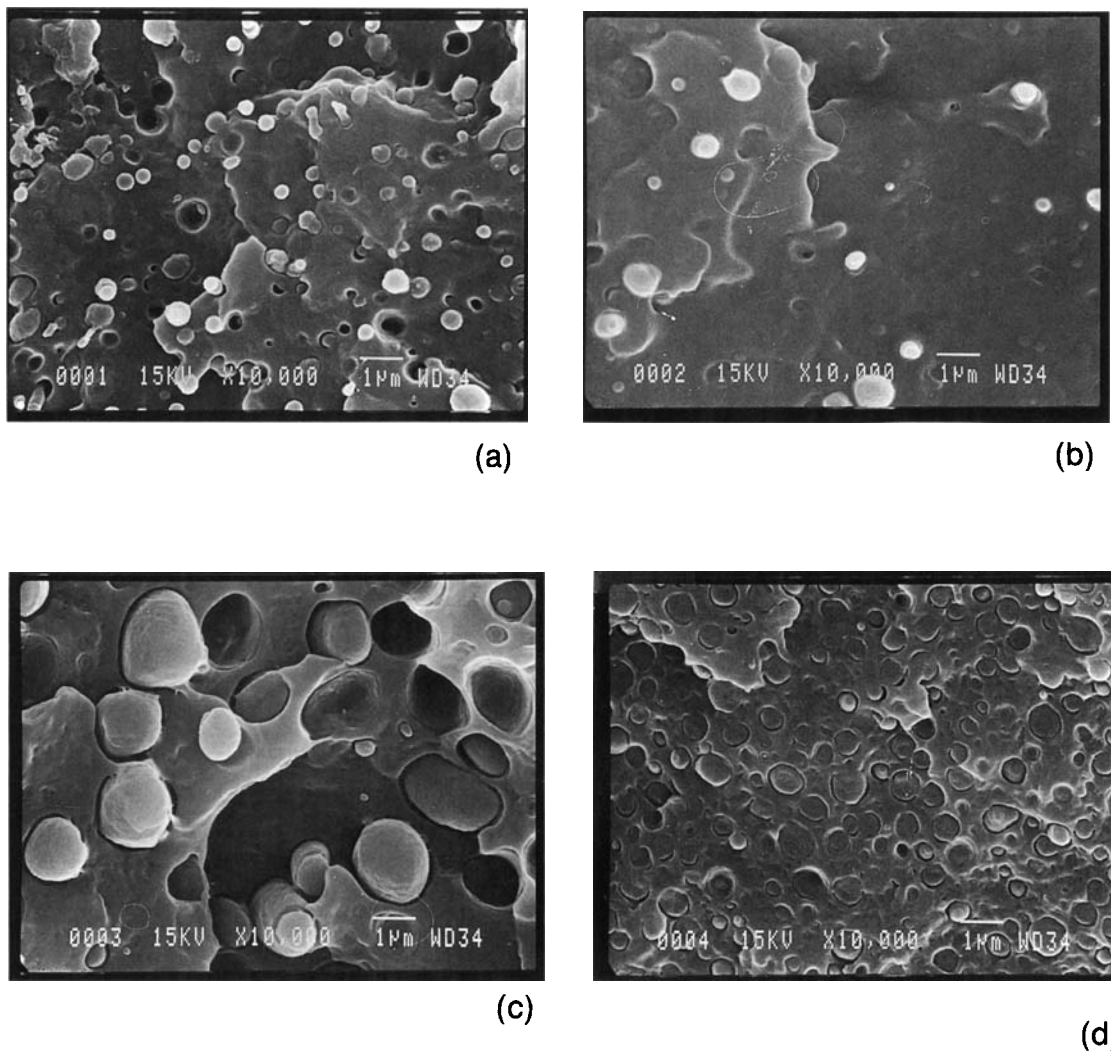


Figure 9 SEM micrographs of the fractured surface of PP/PBT 80/20 blend (a) without and (b) with 5 wt % of the compatibilizer, and PP/PBT 20/80 blend (c) without and (d) with 5 wt % of the compatibilizer ($\times 10,000$).

strength and stiffness obtained. At this composition the compatibilizer did not have a significant effect on the morphology or properties, but it perhaps made the LCP fibrils slightly thinner.

CONCLUSIONS

Binary blends of a reactive ethylene-based terpolymer with polybutylene terephthalate (PBT) and with a liquid crystalline polyester (LCP) were studied by a number of methods to clarify possible interactions between the blended polymers. The aim was to study the suitability of a reactive terpolymer containing epoxy reactivity as a compatibilizer in

blends of polypropylene (PP) and these two polyesters.

Study of the binary blends revealed that chemical reactions take place between the compatibilizer and both PBT and LCP. Probably the carboxyl end groups of the polyesters react with the epoxy group of the compatibilizer. Increased viscosity during blending, changes in the crystallization of the PBT phase, and the intimate contact between the blended polymers found in SEM micrographs pointed to strong interactions or chemical reactions. Torque increased during mixing as a consequence of the reactions and depended on time, temperature, and shear rate. FTIR analysis confirmed the reaction of the epoxide and formation of new esters that were

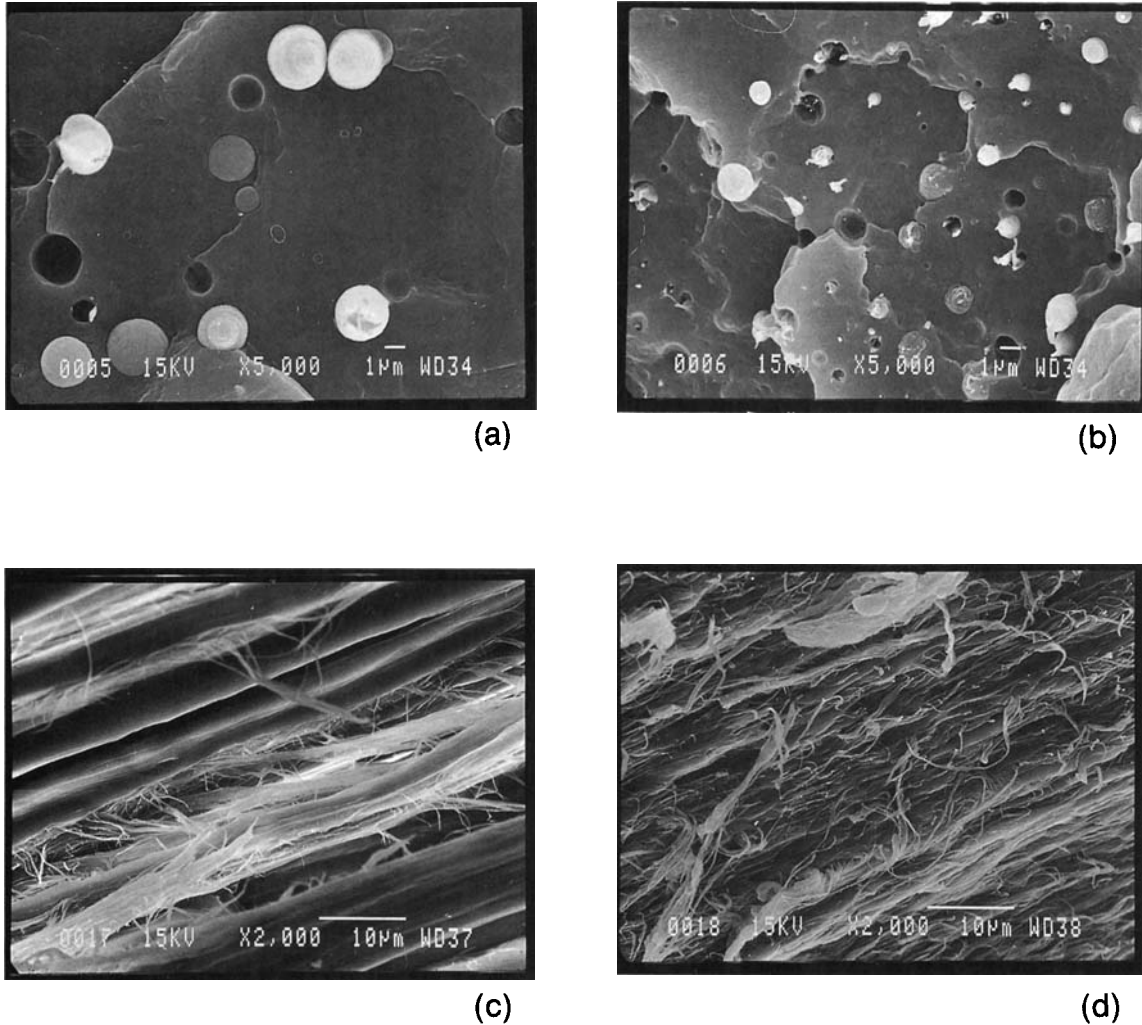


Figure 10 SEM micrographs of the fractured surfaces of compatibilized and noncompatibilized PP/LCP blends: PP/LCP 80/20 blend (a) without and (b) with 5 wt % of the compatibilizer ($\times 5,000$), and longitudinally fractured PP/LCP 20/80 blend (c) without and (d) with 5 wt % compatibilizer ($\times 2,000$).

assumed to take place between the compatibilizer and the polyesters.

In the second part of the work the same terpolymer was used as a compatibilizer in PP/PBT and PP/LCP blends. Increased interactions between the blended polymers through compatibilization were observed as an improved impact strength, in particular in PP/PBT blends, and as significant changes in morphology of both blend systems. Compatibilized PP/PBT blends exhibited synergistic behavior in impact strength measured for notched samples at compositions of 80/20 and 20/80 exceeding the values of both pure PBT and pure PP. The synergism was based on a very fine dispersion of the minor phase well embedded in the matrix.

In PP/LCP blends the compatibilizer significantly improved the impact strength of unnotched samples at the lowest LCP content (20 wt %). Again, it dispersed the LCP phase to small domains well attached to the PP matrix. At higher contents, however, it had a much weaker effect on impact strength. Owing to the softness of the compatibilizer, the strength and stiffness of the blends were slightly reduced in the compatibilized blends. Smaller amounts of the compatibilizer than used here (5 wt %), if well dispersed to the melt, could have similar compatibilizing effect without decrease in other properties.

This study shows that the epoxy functionalized ethylene-based terpolymer acts as a compatibilizer

in both PP/PBT and PP/LCP blends. This behavior is based on good mixing with the PP phase and, as evidenced through investigations on binary blends, chemical reactivity or strong interactions with the polyesters.

As shown, the blend composition and content of the compatibilizer relative to the major blend components are important determinants of the blend properties. Not only the interfacial adhesion induced by the compatibilizer but the viscosity ratio of the blended polymers and the blending and processing conditions are important parameters in determining the blend morphology and final properties. Different ways of feeding the polymers or of preblending the compatibilizer with either of the blend components may be critical to the morphology, as well.

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