

# Properties of Ternary *In Situ* Polycarbonate/Polybutylene Terephthalate/Liquid Crystalline Polymer Composites

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**ABSTRACT:** Ternary *in situ* polycarbonate (PC)/polybutylene terephthalate (PBT)/liquid crystalline polymer (LCP) composites were prepared by injection molding. The liquid crystalline polymer used was a versatile Vectra A950. The matrix of composite was composed of PC/PBT 60/40 by weight. A solid epoxy resin (bisphenol type-A) was used as a compatibilizer for the composites. Dynamic mechanical analysis (DMA) showed that epoxy resin was effective to improve the compatibility between PC and PBT, and between PC/PBT and LCP, respectively. Tensile tests revealed that the stiffness of composites shows little change with the LCP content up to 10 wt %. Above this concentration, the stiffness tended to increase with increasing LCP content. Furthermore, the tensile strengths appeared to increase with increasing LCP content, and their values were close to those predicted from the rule of mixtures. Scanning electron microscopic examination showed that LCP ribbons and short fibrils were developed in the composites containing LCP content  $\leq 10$  wt %. However, fine and elongated fibrils were formed in the skin and core sections of the composites when the LCP content reached 25 wt % and above. Thermogravimetric analysis indicated that the thermooxidative stability of the PC/PBT 60/40 blend tended to improve with increasing LCP content. © 1999 John Wiley & Sons, Inc. *J Appl Polym Sci* 74: 1827–1835, 1999

**Key words:** polycarbonate; polybutylene terephthalate; liquid crystalline polymer; injection molding

## INTRODUCTION

Polycarbonate (PC) and poly(butylene terephthalate) (PBT) are important commercial engineering polymers, each of them providing superior performance in a variety of applications. PC is an amorphous polymer and characterized by its dimensional stability, transparency, flame resistance, high impact strength, and a very wide range of service temperature. However, PC has some shortcomings, such as poor solvent resistance, low fatigue strength, and high melt viscosity. PBT is a

semicrystalline polymer with good chemical resistance, electric insulation, and processability, and it finds wide applications in the automotive and electronic industries. The PC/PBT blends can inherit the toughness of PC and the chemical resistance of PBT, and are available commercially. Blends of PC and PBT have been studied extensively in recent years. However, conflicting results have been reported in literature regarding their apparent state of miscibility between the blend components, i.e., from complete immiscibility when cast from the solvents to partial immiscibility when melt blended.<sup>1–5</sup>

Recently, there has been considerable interest in the research of polyblends of thermoplastics and liquid crystalline polymers (LCPs).<sup>6–11</sup> This is because LCPs exhibit superior physical proper-

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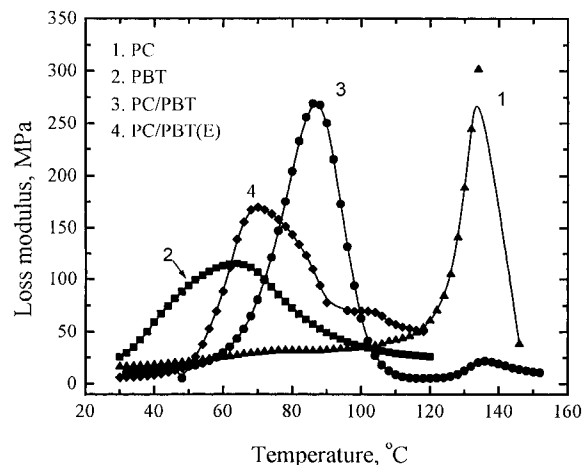
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ties such as low melt viscosity, high stiffness, and high strength. LCPs are composed of long chain rod-like molecules that exhibit an ordered structure in the molten state. The LCPs can develop a fibrillar morphology under certain processing conditions, thereby producing *in situ* composite reinforcement. The structure and mechanical properties of extruded PC/LCP blends are well documented, and commercially available liquid crystalline copolyester (Vectra) is commonly used to reinforce PC matrix.<sup>12–15</sup> Relatively few studies have been conducted on the injection molded PC/LCP blends.<sup>16,17</sup> As PC and Vectra are immiscible, the interfacial bonding between them is relative poor. The degree of LCP fibrillation depends on the LCP content and processing conditions. In general, skin–core structure can develop in both extruded and injection molded PC/LCP blends.<sup>15,17</sup> This means that the LCP domains are deformed into elongated fibrils in the skin layer while they remain as spherical droplets in the core section. For PBT/LCP(Vectra) blends, Heino and Seppala indicated that LCP acts as mechanical reinforcement for PBT, and improves its dimensional and thermal stability.<sup>18</sup> Beery et al. reported that the fibrillation of LCP phase in the extruded PBT/LCP blends depends on the shear rates owing to the viscosity of PBT being smaller than that of LCP. At low shear rates, the LCP phase is mostly spherical, while at higher shear rates the LCP are deformed into short fibrils.<sup>19</sup>

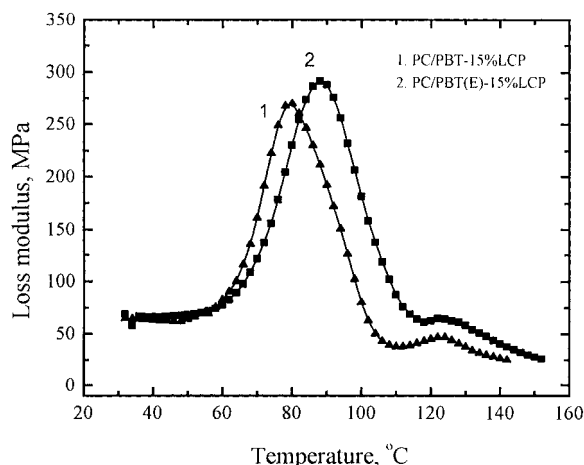
Compared to the large number of studies on LCP/thermoplastic blends, little information is available on the LCP reinforced composites with their matrices consisting of two thermoplastic materials. Generally, these thermoplastics are miscible or partially miscible polymers.<sup>20–22</sup> Bretas and Baird have studied the miscibility and mechanical properties of the ternary poly(ether imide)/poly(ether ether ketone)/LCP blends: the matrices of the *in situ* composites, i.e., PEI/PEEK blends, are miscible at all compositions. They indicated that high modulus can be obtained in ternary blends at high LCP loadings, while compositions with high ultimate tensile strength can be obtained with high loadings of PEI or PEEK.<sup>20</sup> Kwon and Chung have studied the mechanical properties of the extruded blends of PC/poly(ethylene terephthalate) (PET)/LCP.<sup>21</sup> Their results showed that the mechanical properties of PC/PET blends can be enhanced by the addition of less than 10% LCP (Vectra A950). For LCP loadings above 10%, the ternary blends had poor mechanical properties owing to the aggregation of LCP in



**Figure 1** Loss modulus spectra for PC, PBT, and PC/PBT and PC/PBT(E) specimens.

the PC/PET matrix.<sup>21</sup> More recently, Xu et al.<sup>22</sup> have prepared ternary PC/PBT/LCP *in situ* composites by one-step injection molding of LCP pellets with PC and PBT. They used liquid-crystal poly(ester-co-amide) (Vectra B950), which contains terephthalic acid as the reinforcement material. They indicated that addition of PBT to a PC matrix improves the fibrillation of LCP in the matrix and also enhances the adhesion between the matrix and LCP. Consequently, the moduli of ternary PC/PBT/LCP composites are higher than those of PC/LCP blends when LCP content is more than 10%.<sup>22</sup>

Taking advantage of the positive aspects of PC/PBT blends mentioned above, the incorporation of LCP phase into the PC/PBT blends can further enhance their mechanical properties and thermal stability. In this study, we attempt to use a versatile Vectra A950 as the reinforcement polymer for PC/PBT blends. As Vectra A950 is incompatible with PC and PBT, a low-cost solid epoxy resin is used to improve the compatibility between the LCP and thermoplastic components. This solid epoxy has been reported to be an efficient reactive compatibilizer for the PC/polyamide-6 blends and PET/LCP composites.<sup>23</sup> Another study, carried out by Holsti-Mittinen et al.,<sup>7</sup> also showed that a reactive epoxy can be used to compatibilize the polypropylene/PBT and PP/LCP blends. This is because strong interactions or chemical reactions occur between the epoxy and both polyesters.<sup>7</sup> The aim of this article is to investigate the compatibilizing effect of epoxy resin (bisphenol A type) on the morphology and mechanical properties of ternary PC/PBT/LCP blends.



**Figure 2** Loss modulus spectra for PC/PBT/15%LCP and PC/PBT(E)/15%LCP blends.

## EXPERIMENTAL

### Materials

The LCP used in this work was Vectra A950 produced by Hoechst Celanese Company. The composition of this LCP consisted of 27 mol % of 2,6 hydroxynaphthoic acid (HNA) and 73 mol % of *p*-hydroxybenzoic acid (HBA). PC (Makrolon 2606) was supplied by Bayer Company, and PBT (Type 1401) was purchased from Toray Industry, Inc. Solid-state epoxy resin (NPES-909) with an epoxide equivalent weight of 2389 g/equiv. was generously supplied by Nan Ya Plastics of Taiwan.

### Blending Procedure

The PC, PBT, and LCP pellets were dried in an oven at 100°C prior to blending. The PC/PBT 60/40 blend was selected as the matrix of composites. The matrix was initially prepared in a twin-screw Brabender Plasticorder at 265°C and 30

rpm. The extrudates were subsequently pelletized. Similarly, the compatibilized PC/PBT/Epoxy matrix (PC/PBT(E)) was extruded in the Brabender at 265°C and 40 rpm. The blending was performed by mixing 100 parts per hundred (phr) of PC/PBT 60/40 pellets with 2 phr epoxy resin. The extrudates were also pelletized. The matrix and LCP pellets were mixed thoroughly in a plastic box prior to injection molding. Dog bone-shaped tensile bars (ASTM D-638) were injection molded directly from these mixed pellets. The LCP contents of PC/PBT(E)/LCP blends were fixed at 5, 10, 15, 25, and 35%. The barrel zone temperatures of injection molder were set at 285, 290, and 285°C.

### Mechanical Measurements

The tensile behavior of the blends was determined using an Instron tensile tester (model 4206) at room temperature under a crosshead speed of 1 mm/min. The gauge length of specimens was 57 mm. At least five specimens of each composition was tested and the average values reported.

Izod impact specimens with dimensions of 63 × 13 × 3.2 mm were cut from the midsection of the tensile bars. They were tested using a Ceast impact pendulum tester. At least seven specimens were tested and the average values reported.

### Morphological Observation

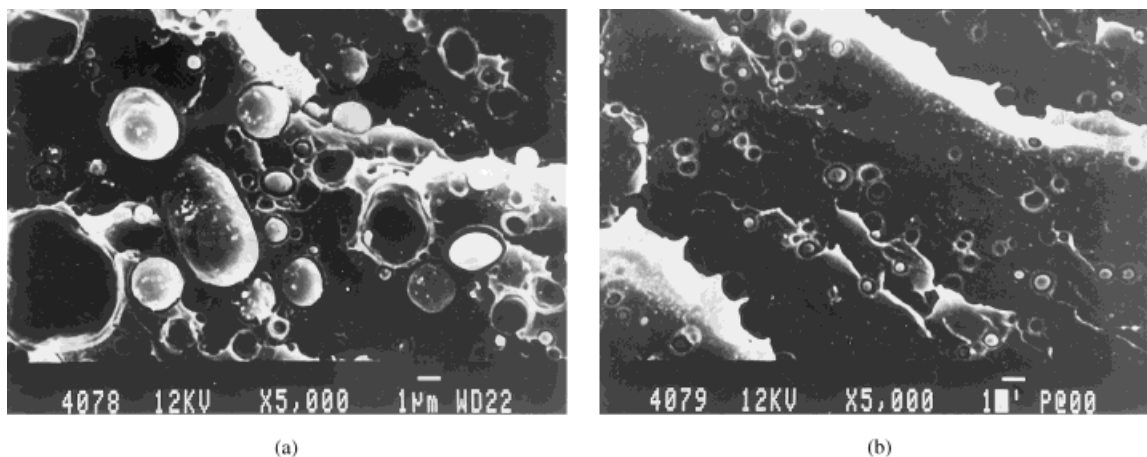
The morphologies of the fractured surfaces of the blends were observed in a scanning electron microscope (SEM; JEOL JSM 820). The blend specimens were cryo-fractured in liquid nitrogen. They were then coated with a thin layer of gold prior to SEM observations.

### Dynamic Mechanical Analysis

Dynamic mechanical analysis (DMA) of the injection molded specimens were conducted using a

**Table I** Glass Transition Temperatures for PC, PBT, and Their Blends

	$T_g^{\text{PC}}, ^\circ\text{C}$	$T_g^{\text{PBT}}, ^\circ\text{C}$	$T_g^{\text{LCP}}, ^\circ\text{C}$	$D = \frac{(T_g^{\text{PC}} \text{ or } T_g^{\text{LCP}})}{T_g^{\text{PBT}}}$
PC	136.1	—	—	—
PBT	—	62.8	—	—
PC/PBT 60/40	136.4	85.6	—	50.8
PC/PBT (E) 60/40	100.2	70.0	—	30.2
PC/PBT/15% LCP	—	80.0	122.3	42.3
PC/PBT (E)/15% LCP	—	88.4	122.1	35.7



**Figure 3** SEM micrographs of (a) PC/PBT and (b) PC/PBT(E) blends.

Du Pont dynamic mechanical analyzer (model 980) at a fixed frequency of 1 Hz and an oscillation amplitude of 0.4 mm. The temperature range studied was from  $-30$  to  $150^{\circ}\text{C}$ , with a heating rate of  $4^{\circ}\text{C}/\text{min}$ .

#### Thermogravimetric Analysis (TGA)

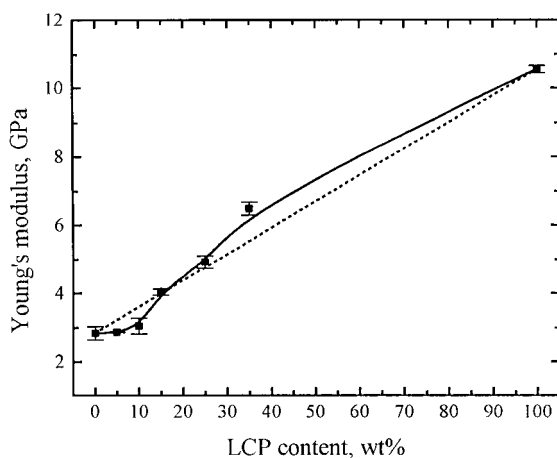
The decomposition process of the blend specimens from  $30$ – $600^{\circ}\text{C}$  under a protective helium atmosphere ( $200$  mL/min) was determined with a thermogravimetric analyzer (Seiko model SSC/5200). The heating rate employed was  $10^{\circ}\text{C}/\text{min}$ .

## RESULTS AND DISCUSSION

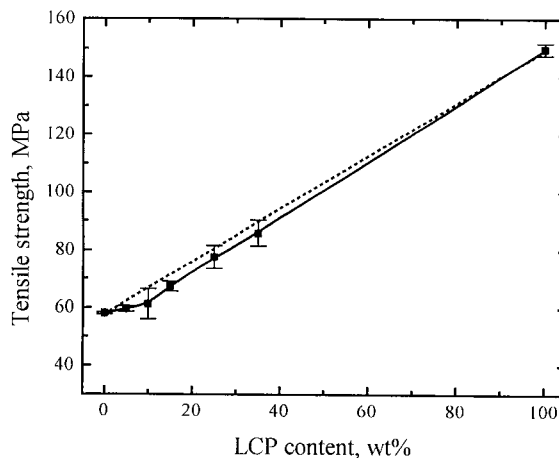
### Compatibility

Figure 1 shows the typical loss modulus spectra for the PC, PBT, PC/PBT, and PC/PBT(E) speci-

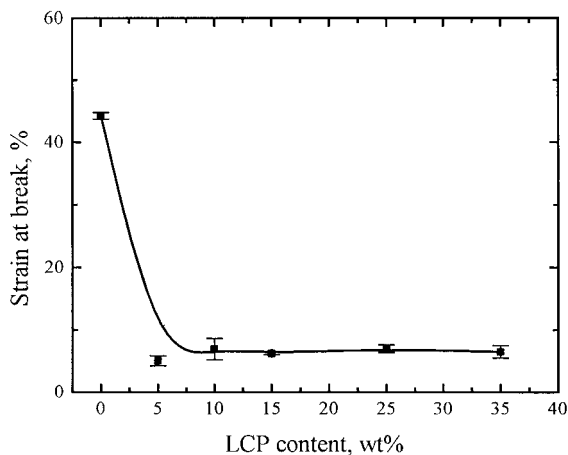
mens. The glass transition temperatures ( $T_g$ ) for pure PC and PBT are  $136$  and  $63^{\circ}\text{C}$ , respectively. Furthermore, it can be seen that the  $T_g$  for the PBT phase of the PC/PBT blends is shifted to  $86^{\circ}\text{C}$ , while the  $T_g$  of the PC phase remains unchanged. The  $T_g$  of  $86^{\circ}\text{C}$  is possibly related to the ester exchange reaction, thereby increasing the extent of miscibility between the PC and PBT phases. Wahrmond et al.<sup>24</sup> have studied the dynamic mechanical behavior of the PC, PBT, and PC/PBT blends. The  $T_g$ s for the pure PC and PBT determined from the loss modulus spectra are  $150$  and  $60^{\circ}\text{C}$ , respectively. The  $T_g$  of the PBT phase of the PC/PBT blend is about  $15^{\circ}\text{C}$  or so higher than that of the pure PBT.<sup>24</sup> From Figure 1, addition of 2 phr epoxy to the PC/PBT(E) blend has resulted in the shift of  $T_g$ s of these two components towards each other. The compatibility between the PC and PBT is improved accordingly. It is noted that the shift in  $T_g$  of the PBT phase of



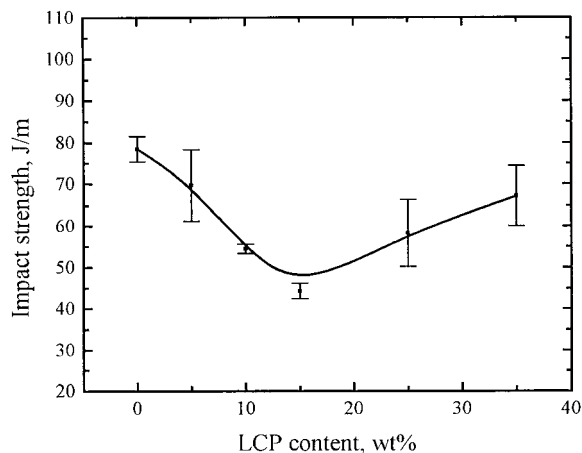
**Figure 4** Variation of Young's modulus with LCP content for ternary *in situ* composites.



**Figure 5** Variation of tensile strength with LCP content for ternary *in situ* composites.



**Figure 6** Variation of strain at break with LCP content for ternary *in situ* composites.

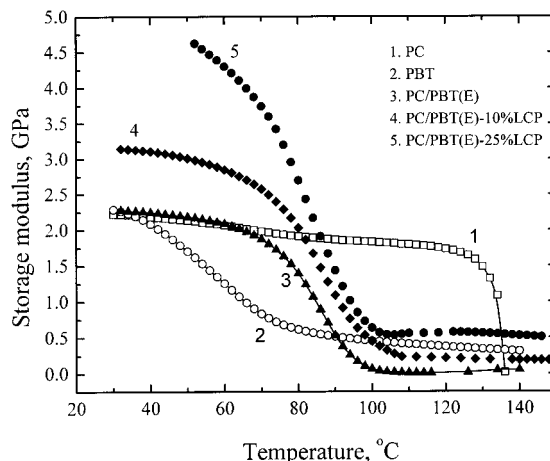


**Figure 7** Variation of Izod impact strength with LCP content for ternary *in situ* composites.

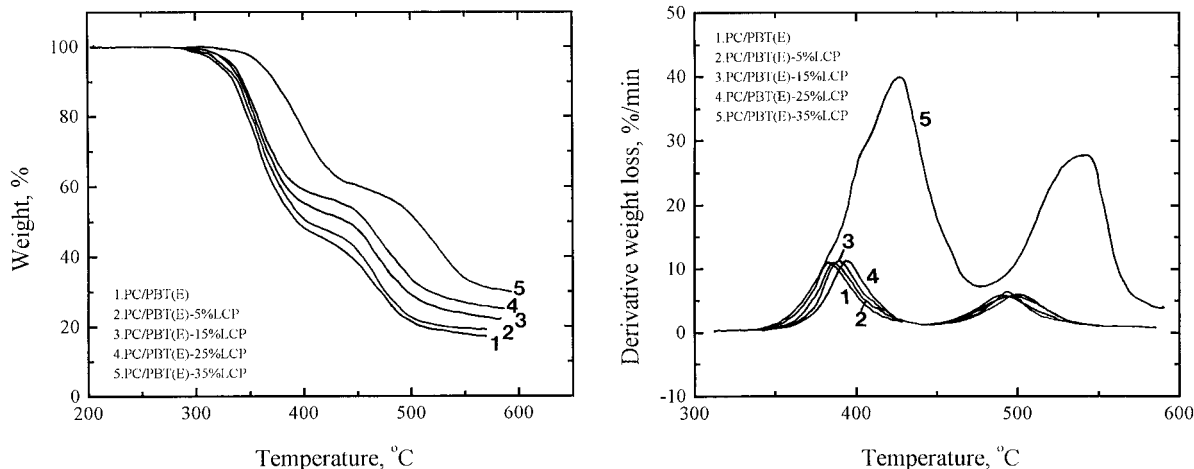
PC/PBT(E) 60/40 blend is smaller than that of the PC/PBT blend, indicating that the epoxy resin may suppress the exchange reaction in the PBT-rich phase. Figure 2 shows the loss modulus spectra for the PC/PBT/15%LCP and PC/PBT(E)/15%LCP blends. It is evident that the epoxy resin addition is also beneficial to improve the compatibility between the LCP and PC/PBT matrix. The  $T_g$ s of several specimens are summarized in Table I. In this table, the difference in  $T_g$  values (D) between the blend components are also tabulated. It is apparent that the D values of the compatibilized PC/PBT(E) and PC/PBT(E)/LCP blends are smaller than their counterparts.

The epoxy addition can also lead to a microstructural change in the PC/PBT blend. Figure 3(a) shows the SEM micrograph of the uncompatibilized PC/PBT blend. It can be seen that the PBT disperses into large domains in a PC-rich matrix. Some PBT domains have a size of up to  $\sim 6 \mu\text{m}$ . Although the ester reaction takes place in this blend, it is considered that its effect on the compatibility between PC and PBT is very small, due to the presence of large-sized PBT-rich domains. However, the epoxy resin addition gives rise to a finer distribution of PBT in the PC matrix. The size of the PBT domains is reduced to  $\sim 0.5 \mu\text{m}$  [Fig. 3(b)]. On the basis of dynamic and SEM examinations, it is evident that the epoxy resin can react with the end groups of PC, PBT, and LCP, i.e.,  $-\text{COOH}$  or  $-\text{OH}$ , thereby facilitating the formation of the block copolymer. Such an *in situ* copolymer appears to improve the compatibility between PC and PBT, and between PC/PBT and LCP considerably. According to the litera-

ture, solid-state epoxy is very effective to compatibilize the polymer blends containing PBT as a blend component, for example, polyamide 66/PBT blends. This is because epoxy tends to react with the aliphatic hydroxyl ( $-\text{OH}$ ) or carboxyl ( $-\text{COOH}$ ) end groups of PBT, thereby forming an epoxy-*b*-PBT copolymer at the interface. This mixed copolymer is believed to be the major contributor in improving the compatibility of PA66/PBT blends. Similarly, Holsti-Miettinen et al.<sup>7</sup> also indicated that the epoxy-functionalized polymer improves the impact strength of the PP/PBT and PP/LCP blends, owing to the fact that the epoxy group of the compatibilizer reacts with the carboxyl end groups of the polyesters. In both blends, the compatibilizer reduces the size of dis-



**Figure 8** Storage modulus vs. temperature for PC, PBT, and their blends.



**Figure 9** (a) TG and (b) DTG curves for PC/PBT(E) blend and ternary composites.

persed domains and causes them to attach better to the matrix.<sup>7</sup>

### Mechanical Behavior

For the uncompatibilized PC/PBT 60/40 blend, static tensile tests show that the tensile strength, stiffness, and strain at break for this specimen are 60 MPa, 2.8 GPa, and 30%, respectively. The impact energy for this blend is 55 J/m. Addition of 2 phr epoxy to the PC/PBT 60/40 blend leads to a minute change in the tensile strength and stiffness, but to a considerable increase in strain at break (44%) and impact energy (79 J/m). Thus, our attention is mainly paid to the compatibilized PC/PBT(E) and PC/PBT(E)/LCP blends. Figure 4 shows the variation of Young's modulus, with the LCP content for the ternary composites investigated along the flow direction. It is noticed that the addition of the LCP content  $\leq 10$  wt % has little effect to increase the stiffness of the composites. Above 10 wt %, the stiffness of composites tends to increase with increasing the LCP content, and deviates positively from the rule of mixtures. It is worth mentioning that the rule of mixtures is generally expressed in volume percentage rather than the weight percentage. The low stiffness for the composites containing LCP  $\leq 10$  wt % is due to the formation of short LCP fibrils in the skin layer, as will be discussed in the next section. The tensile strength vs. LCP content for the composites investigated is shown in Figure 5. Apparently the tensile strengths of the composites are very close to those predicted from the rule of mixtures. The variation of strain at break vs. LCP content for the ternary composites is shown

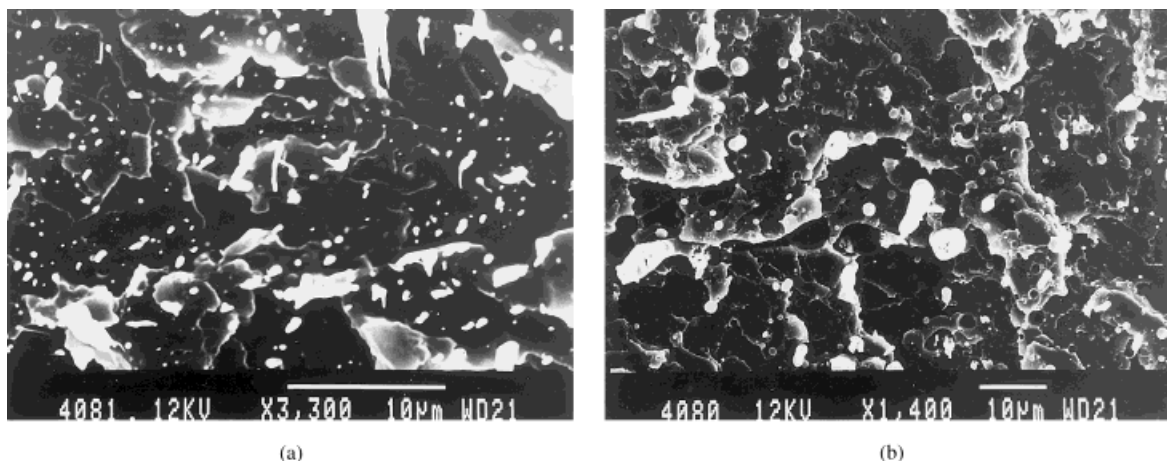
in Figure 6. The strain at break decreases dramatically with increasing LCP content as expected, and this is a typical characteristic of the polymer composites.

Figure 7 shows the Izod impact strength vs. the LCP content for the PC/PBT(E)/LCP composites. This figure reveals that the impact strength initially decreases with increasing the LCP content up to 15 wt %; thereafter, it tends to increase with increasing the LCP content. In previous studies, we have observed a similar variation trend of impact strength with the LCP content for the maleic anhydride (MA) compatibilized PP/LCP and polyamide 6/LCP composites.<sup>10,25</sup> This is attributed to the MA-g-PP acting as an effective compatibilizer for these blends, thereby yielding long LCP fibrils in the matrix at higher LCP loadings. Furthermore, the improved impact strength originates from the enhanced matrix plasticity at fibril ends in the crack tip region when the LCP fibrils are sufficiently close spaced.

Figure 8 shows the representative dynamic mechanical curves that show the variation of stor-

**Table II** Thermal Properties for PC/PBT (E)/LCP Composites

Specimen	$T_{-5\%}$ , °C	$T_{\max}^1$ , °C	$T_{\max}^2$ , °C
PC/PBT (E)	360.8	384.4	—
PC/PBT (E)/5% LCP	361.5	384.5	483.8
PC/PBT (E)/10% LCP	363.3	386.7	490.0
PC/PBT (E)/15% LCP	365.3	387.1	490.5
PC/PBT (E)/25% LCP	368.3	388.3	493.0
PC/PBT (E)/35% LCP	391.6	423.5	541.1



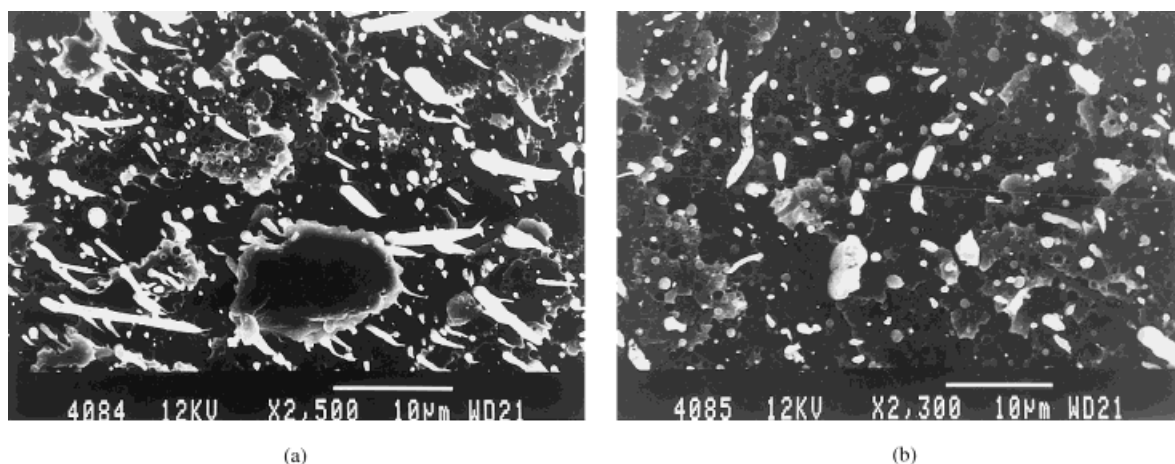
**Figure 10** SEM fractographs of (a) skin and (b) core sections of the injection-molded PC/PBT(E)/10% LCP composite.

age modulus with temperature for PC, PBT, and their blends. The storage modulus of PC shows little change from 30 to 120°C, but it drops significantly at  $T_g$ . This is a typical behavior of amorphous polymers. This behavior enables PC to be used safely for a wide range of service temperatures. In contrast, the storage modulus of PBT decreases continuously with increasing temperatures. The incorporation of PC into PBT appears to stabilize the storage modulus of PBT considerably up to about 70°C. Thus, the PC/PBT(E) blend inherits the good thermal stability of PC. It is noticed from Figure 8 that the stability and value of the storage modulus of the PC/PBT(E) blend can be enhanced further by adding LCP at  $\geq 10$  wt %. Finally, the storage moduli of uncompatibilized PC/PBT and PC/PBT-LCP blends remain

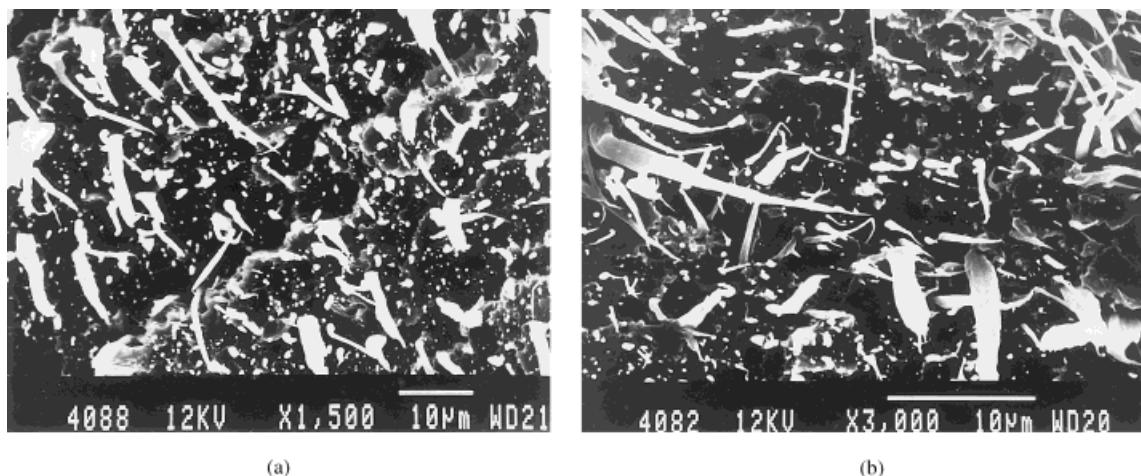
nearly unchanged. Thus, the solid-state epoxy is only beneficial to improve the impact toughness of the blends, and it has little effect on increasing the tensile strength and stiffness of the blends investigated.

#### Thermogravimetric Analysis

Figure 9(a)–(b) shows the thermogravimetric (TG) and derivative weight loss (DTG) curves for the PC/PBT(E) blend and ternary composites investigated. The 5% weight loss temperature ( $T_{-5\%}$ ) determined from the  $T_g$  curves and the maximum weight loss temperature ( $T_{max}$ ) determined from the DTG curves are summarized in



**Figure 11** SEM fractographs of (a) skin and (b) core sections of the injection-molded PC/PBT(E)/15% LCP composite.



**Figure 12** SEM fractographs of (a) skin and (b) core sections of the injection-molded PC/PBT(E)/25% LCP composite.

Table II. In this table,  $T_{\max}^1$  corresponds to the maximum weight loss of the PC/PBT(E) matrix, where  $T_{\max}^2$  is associated with that of the LCP phase. These data indicate that the  $T_{-5\%}$  and  $T_{\max}$  tend to increase with increasing the LCP content. Hence, LCP addition is beneficial to improve the thermooxidative stability of the PC/PBT blend.

#### LCP Fibrillation

Figure 10(a)–(b) shows SEM micrographs of the skin and core sections of the PC/PBT(E)/10%LCP blend. It can be seen from Figure 10(a) that the LCP phase deforms mainly into ribbons of the PC/PBT(E) matrix. In the core section, the LCP phase disperses as ellipsoids and spherical droplets. As the LCP content in the ternary blend is increased to 15 wt %, the LCP phase begins to deform into short fibrils in the skin and core sections [Fig. 11(a)–(b)]. However, fine and elongated LCP fibrils tend to develop in the skin and core sections of the composites when the LCP content reaches 25 wt % [Fig. 12(a)–(b)]. In this case, the stiffness of the composites improves dramatically when the LCP content was  $\geq 25$  wt % (Fig. 4). It is well reported in the literature that LCP fibrils are rather difficult to form in the PC/LCP blends, and the fibrillation depends on the processing conditions and LCP concentration.<sup>12–17</sup> However, the incorporation of PBT improves the processability of PC, thereby facilitating the LCP fibrillation.<sup>22</sup> In this study, the epoxy resin improves the compatibility between the PC and PBT, and between the PC/PBT and LCP effectively, leading to the

formation of elongated fibrils in ternary composites at LCP loadings of 25 wt % and above.

#### CONCLUSION

Ternary PC/PBT/LCP blends were prepared by injection molding. Bisphenol A epoxy resin was found to improve the compatibility between the blend components effectively. Static tensile tests showed that the stiffness of the ternary blends showed little change with the addition of the LCP content of  $\leq 10$  wt %. Above 10 wt %, the stiffness tended to increase with increasing the LCP content. Moreover, the tensile strengths of ternary blends appeared to increase with increasing the LCP content, and their values were very close to those predicted from the rule of mixtures. Izod impact test showed that the impact strength of *in situ* composites initially decreased with increasing the LCP content up to 15 wt %; thereafter, it showed an increase with increasing the LCP content. SEM observations revealed that the epoxy resin leads to a finer dispersion of PBT in the PC matrix of a binary PC/PBT blend. For ternary PC/PBT(E)/LCP blends with low LCP loadings, the LCP phase was dispersed as ribbons and short fibrils in the skin section of the specimens. However, elongated LCP fibrils developed in both the skin and core section of the ternary blends when the LCP content were increased to 25 wt % and above. Finally, LCP addition was beneficial to improve the thermooxidative stability of the PC/PBT 60/40 blend.



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