

Mechanical Performance of Ternary *In Situ* Polycarbonate/Poly(acrylonitrile-butadiene-styrene)/Liquid Crystalline Polymer Composites

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ABSTRACT: Ternary *in situ* polycarbonate (PC)/poly(acrylonitrile-butadiene-styrene) (ABS)/liquid crystalline polymer(LCP) composites were prepared by injection molding. The LCP used was a versatile Vectra A950, and the matrix of composite specimens was PC/ABS 60/40 by weight. Maleic anhydride (MA) copolymer and solid epoxy resin (bisphenol type-A) were used as compatibilizers for these composites. The tensile, dynamic mechanical, impact, morphology, and thermal properties of the composites were studied. Tensile tests showed that the tensile strength of the PC/ABS/LCP composite in the longitudinal direction increased markedly with increasing LCP content. However, it decreased slowly with increasing LCP content in the transverse direction. The modulus of this composite in the longitudinal direction appeared to increase considerably with increasing LCP content, whereas the incorporation of LCP into PC/ABS blends had little effect on the modulus in the transverse direction. The impact tests revealed that the Izod impact strength of the composites in both longitudinal and transverse direction decreased with increasing LCP content up to 15 wt %; thereafter it increased slowly with increasing LCP. Dynamic mechanical analyses (DMA) and thermogravimetric measurements showed that the heat resistance and heat stability of the composites tended to increase with increasing LCP content. Scanning electron microscopy observation and DMA measurement indicated that the additions of epoxy and MA copolymer to PC/ABS matrix appeared to enhance the compatibility between the PC and ABS, and between the matrix and LCP. © 1999 John Wiley & Sons, Inc. *J Appl Polym Sci* 74: 2274–2282, 1999

Key words: compatibilizer; polycarbonate; ABS; epoxy; tensile strength

INTRODUCTION

Polycarbonate (PC) is a widely used engineering thermoplastic because it possesses several distinct properties such as transparency, dimensional stability, flame resistance, high heat distortion temperature, and high impact strength. However, PC is known to exhibit high notch sensitivity, and is susceptible to crazing or cracking

on exposure to various solvents. Moreover, PC is relatively soft and the surface of polymer can be easily scratched.¹ Acrylonitrile-butadiene-styrene (ABS) copolymer is a most successful rubber-toughened thermoplastic that has several attractive properties such as good processability, notch insensitivity, and low cost. So ABS plastic is used as a material for the golf-ball cover, protective helmet, telephone switchboard panel, screw driver handle, etc.² It also finds applications in the automotive industry because its surface finishes are relatively smooth and can be easily decorated. However, ABS has certain limitations,

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namely low thermal stability, and poor flame and chemical resistance.

Fabrication of multiphase plastics generally offers a route to combine several good properties of the individual polymer components. It can be achieved by mixing two or more polymers or by filling polymers with inorganic particulates or fibrous fillers, leading to the formation of polymer blends or composites.³⁻⁵ In this context, the combination of the advantageous properties of ABS and PC has resulted in the development of commercially available PC/ABS blends that are useful in many molding applications, particularly in the automotive industry.⁶ In general, the mechanical properties and morphology of PC/ABS blends depend on the PC molecular weight, processing conditions, type, and content of PC, and the compatibility between ABS and PC.⁶⁻¹³ In the latter case, there has been much interest recently on the topics concerned with the improvement of the mechanical performances of ABS/PC blends by adding functionalized additives in order to reduce the interfacial tension, and to promote the adhesion at the interface. For example, Carrot et al. have successfully grafted maleic anhydride (MA) onto ABS in the molten state, and the resulting MA-g-ABS copolymer can react with polymers containing reactive end groups.¹⁴ Ning and Ko indicated that MA-grafted polypropylene (MAP) can act as an effective compatibilizer or a processing agent for PC.¹⁵ More recently, Jin et al reported that poly(methyl methacrylate) can be used to enhance the compatibility between ABS and PC.¹⁶

Recently, research of polyblends of thermoplastics and liquid crystalline polymers (LCPs) has attracted considerable attention of polymer scientists.¹⁷⁻²³ This is because LCPs exhibit superior physical properties 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. When small amounts of LCPs are blended with thermoplastics, the LCPs can deform into fine fibrils under certain processing conditions, leading to the formation of *in situ* composites. The degree of LCP fibrillation depends on the LCP content and processing conditions.

Although there are numerous publications on the LCP/thermoplastic blends, less information is available on LCP-reinforced composites with matrices consisting of two thermoplastic materials. Generally, these thermoplastics are miscible or partially miscible polymers.²⁴⁻²⁶ For example,

Bretas and Baird have studied the miscibility and mechanical properties of ternary poly(ether imide)(PEI)/poly(ether ether ketone)(PEEK)/LCP blends. The matrices of this *in situ* composite are PEI/PEEK blends, and are miscible at all compositions. The authors reported that high stiffness can be achieved in ternary blends containing high LCP content, while high ultimate tensile strength can be obtained with high loadings of PEI or PEEK.²⁴ Recently, Kwon and Chung have studied the mechanical properties of extruded blends of PC/poly(ethylene terephthalate) (PET)/LCP.²⁵ Their results showed that the mechanical properties of PC/PET blends can be enhanced by the addition of less than 10% LCP. For LCP loadings above 10%, the ternary blends exhibited poor mechanical properties owing to the aggregation of LCP in the PC/PET matrix. Xu et al.²⁶ have prepared ternary PC/PBT/LCP *in situ* composites by one-step injection molding of LCP pellets with PC and PBT. They indicated that the moduli of ternary PC/PBT/LCP composites are higher than those of PC/LCP blends when the LCP content is higher than 10%.²⁶ It is worth mentioning that the LCPs used by these workers were copolyesters such as HX400, Vectra A950, and Vectra B950.²⁴⁻²⁶ More recently, Tjong and Meng have used a solid epoxy resin (bisphenol A type) to improve the compatibility between PC and PBT, and between a PC/PBT blend and LCP (Vectra A950), respectively. The results showed that the epoxy resin addition leads to a finer dispersion of PBT in the PC matrix of a binary PC/PBT blend, and to the formation of elongated LCP fibrils in the PC/PBT matrix.²⁷ In this paper, we attempt to use the epoxy resin and MAP to improve the compatibility between the PC and ABS, and between the PC/ABS and LCP. The aim is to enhance the mechanical strength and thermal stability of the PC/ABS composites containing LCP.

EXPERIMENTAL

Materials

The PC was produced by Bayer Company (Germany) under the trade name of Makrolon 2605, while ABS (type 100) used in this study was purchased from Toray plastics (Malaysia). The LCP (Vectra A950) was purchased from Hoechst Celanese Company (USA). The composition of this LCP consisted of 27 mol % of 2,6-hydroxynaphthoic acid (HNA) and 73 mol % of *p*-

hydroxybenzoic acid (HBA). MAP (Hercoprime HG 201) was supplied by Himont Company, and the solid epoxy resin (NPES) was kindly given by Nan Ya Plastics of Taiwan.

Sample Preparation

The PC, ABS, and LCP pellets were dried in an oven at 100°C for 24 hr prior to blending. The blend consisting of 95% ABS and 5% MAP by weight was prepared in a twin-screw Brabender Plasticorder at 240°C and 30 rpm. The extrudate was cut into pellets by a pelletizer. These pellets were designated as ABS(M) in this paper. The matrix was prepared by mixing 100 phr PC/ABS(M) 60/40 pellets with 2 phr epoxy resin in Brabender at 245 °C and 30 rpm for about 120 s. Finally, the matrix and LCP pellets were mixed thoroughly in a plastic box prior to injection molding. The plaques with dimensions of 200 × 80 × 3.2 mm were injection molded from these pellets. The LCP contents of the blends were fixed at 5, 10, 15, 25, and 35%. The barrel zone temperatures of the injection molder were set at 285, 290, and 285 °C. These plaques were cut into dog-bone shaped tensile bars. Notched Izod impact specimens were also prepared from the plaques. Both longitudinal and transverse specimens were used for the tensile and impact tests. For the longitudinal specimens, the length direction was parallel to the mold-filling direction, while it was perpendicular to the mold-filling direction for the transverse specimens. In order to study the effect of the additions of compatibilizers on the properties of PC/ABS/LCP composites, both PC/ABS(60/40)/2 phr Epoxy and PC/ABS 60/40 (by weight) composite specimens containing 25 wt % LCP were prepared under similar processing conditions.

Mechanical Measurements

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

Izod impact specimens with dimensions of 65 × 13 × 3.2 mm were cut from the plaques, and were tested by a Ceast impact pendulum tester. These specimens were sharply notched with a V-shape knife. The radius of curvature for the notch is 0.025 mm. At least five specimens were tested and the average values reported.

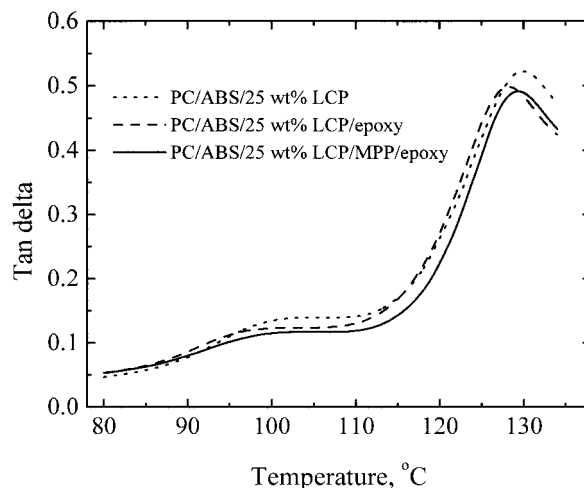


Figure 1 Effect of MAP and epoxy additions on the tan delta spectra of PC/ABS/LCP composites.

Dynamic mechanical analysis (DMA) of the injection molded rectangular specimens with dimensions of 65 × 13 × 3.2 mm were conducted using a Du Pont dynamic mechanical analyzer (model 983) at a fixed frequency of 1 Hz and an oscillation amplitude of 0.4 mm. The temperature studied ranged from 30 to 140°C, and the heating rate employed was 4°C min⁻¹.

Thermal Analyses

Thermal analysis was carried out in a Seiko thermogravimetric analyzer (model SSC/5200). This instrument was also equipped with a differential thermal analyzer. The weight loss against temperature was measured at a rate of 10°C min⁻¹ in a helium atmosphere from 50 to 600°C.

Morphological Observation

The morphologies of the surfaces of the blend specimens were observed in a scanning electron microscope (SEM, Jeol JSM 820). The specimens were cryofractured in liquid nitrogen. All the samples were coated with a thin layer of gold prior to SEM observations.

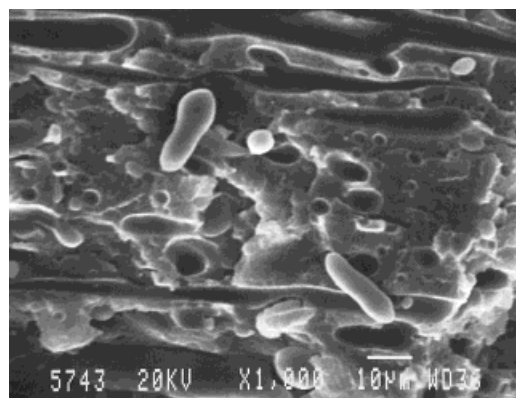
RESULTS AND DISCUSSION

Effects of Compatibilizer

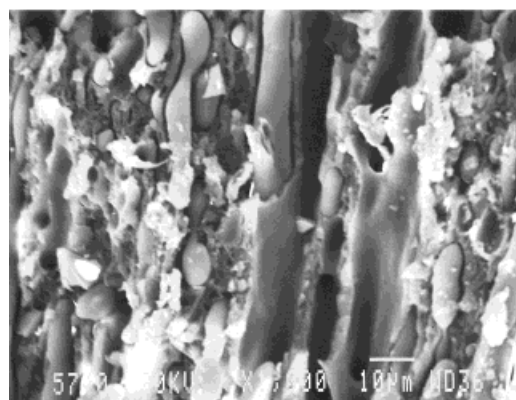
Figure 1 shows the tan delta spectra for the PC/ABS/25wt%LCP, PC/ABS/25wt%LCP/epoxy and PC/ABS(M)/25wt%LCP/epoxy specimens. The glass

transition temperature (T_g) of PC phase of PC/ABS/25 wt % LCP composite is located at 131°C, while the T_g of the ABS phase is roughly at 104°C. Apparently, the addition of MAP and 2 phr epoxy to the composite has resulted in a small shift of the glass transition temperatures of PC and ABS components toward each other. This indicates that the compatibility between the PC and ABS is slightly improved. Moreover, the epoxy addition can also lead to an improvement in the compatibility between LCP and matrix. The shift in T_g s between the PC and ABS phases of compatibilized specimens is relative small as determined by DMA; hence, it is more appropriate to examine the morphology of these specimens. It is generally known that the LCP-reinforced polymer composites exhibit a skin-core morphology. The LCPs tends to deform into microfibrils in the skin layer under appropriate processing conditions, while they disperse as ellipsoids or spherical domains in the core section of in-situ composites. Figures 2(a) and 2(b) show the SEM micrographs of the core sections of the PC/ABS/25 wt % LCP and PC/ABS(M)/25 wt % LCP/epoxy specimens, respectively. Comparing with PC/ABS(M)/25 wt % LCP/epoxy composite, considerable LCP ellipsoids are pulled out from the matrix of the PC/ABS/25 wt % LCP specimen during cryofracture. However, most LCP ellipsoids remain intact with the matrix of the PC/ABS(M)/25 wt % LCP/epoxy specimen. These results indicate that the epoxy resin addition also enhances the interfacial adhesive force between the matrix and LCP. On the basis of DMA and SEM examinations, we can conclude that the MAP and epoxy resin additions improve the compatibility between the PC and ABS, and also between the matrix and LCP.

We now consider the compatibilized effects of MAP and epoxy resin. It is believed that MA end group of ABS(M) can act as a compatibilizer for PC, thereby improving the adhesion between the PC and ABS blend components. According to literature, MAP acts as a compatibilizer and processing agent for PC.¹⁵ Similarly, Tjong and Meng²⁸ reported that the MAP addition can result in a finer dispersion of ABS domains in the PC/ABS blends, leading to marked improvements in tensile ductility and impact strength of the blends. They further pointed out that the incorporation of 2 phr solid epoxy resin into PC/ABS(M) blends renders the ABS domains and PC boundaries become more obscure.²⁸ It is worth noting that solid epoxy resin can also be used to improve the compatibility of the blend compo-



(a)



(b)

Figure 2 SEM fractographs of the core regions for (a) PC/ABS/LCP and (b) PC/ABS(M)/LCP/epoxy composites.

nents of LCP/thermoplastic composites. For example, Chang and co-workers²⁹ indicated that the epoxide end groups of solid epoxy resin is an effective compatibilizer for the incompatible blends of PET and LCP (Vectra A900). This is because solid epoxy resin can react with LCP during the blending process, thereby promoting the formation of the *in situ* epoxy-b-LCP copolymer. Such a copolymer is highly effective in compatibilizing the LCP/PET blends.²⁹

Mechanical Properties

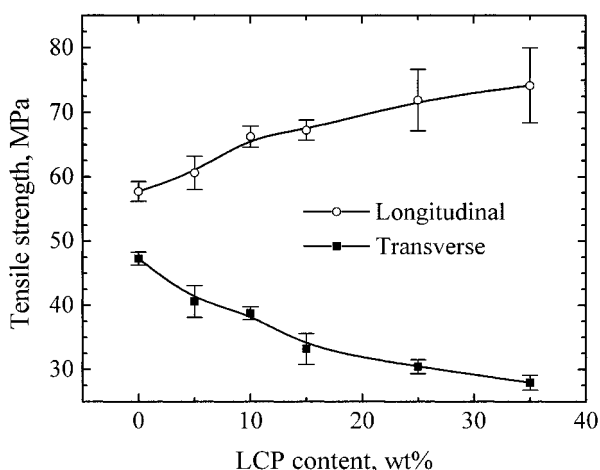
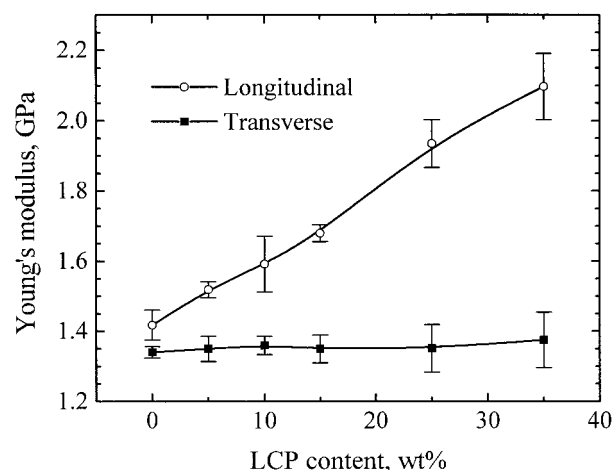
The effects of the compatibilizer additions on the tensile and impact properties of a selected PC/ABS/25wt%LCP blend are listed in Table I. Apparently, the incorporation of bisphenol A type epoxy resin leads to an increase in both the stiffness and tensile strength, but to a slight decrease in impact strength. However, the strain at break

Table I Effects of Compatibilizer Additions on the Longitudinal Mechanical Properties of PC/ABS/25 wt % LCP Blend

Sample	Property			
	Tensile Strength (MPa)	Strain at Break (%)	Young's Modulus (MPa)	Izod Impact Strength (kJ/m ²)
PC/ABS/25 wt % LCP	70 ± 4	4.5 ± 0.4	1907 ± 13	6.9 ± 0.7
PC/ABS/25 wt % LCP/epoxy	76 ± 6	4.7 ± 0.2	2026 ± 78	6.7 ± 0.7
PC/ABS/25 wt % LCP/epoxy/MAP	72 ± 5	4.9 ± 0.3	1935 ± 68	7.7 ± 1.0

and impact strength, particularly the latter, improve considerably with the addition of both MAP and epoxy resin. Thus, we focus our attention on the mechanical and thermal properties of PC/ABS/LCP blends containing MAP and bisphenol A type epoxy resin. The variation of tensile strength with LCP content for PC/ABS(M)/LCP/epoxy samples in both longitudinal and transverse directions is shown in Figure 3. Apparently, the tensile strength of the longitudinal sample is much higher than that of the transverse sample. Moreover, the tensile strength of the longitudinal sample increases considerably with LCP contents. However, the tensile strength of the transverse sample appears to decrease continuously with increasing LCP content. This is a typical behavior of the composites reinforced with LCP.^{30,31} Figure 4 shows the variation of Young's modulus with LCP content for PC/ABS(M)/LCP/

epoxy samples in both longitudinal and transverse direction. In this figure, it can be seen that the stiffness of all longitudinal specimens tends to increase continuously with increasing LCP content, whereas it remains nearly unchanged for the transverse samples. The ratios of tensile strength of the longitudinal sample to those of the transverse samples are shown in Figure 5. In addition, the ratios of stiffness for these specimens are also plotted. It can be seen from this figure that the differences in the mechanical properties between the longitudinal and transverse samples increase markedly with increasing LCP content, especially for the tensile strength. The variation of strain at break with LCP content for PC/ABS(M)/LCP/epoxy samples in both longitudinal and transverse directions is shown in Figure 6. This figure reveals that the strain at break for longitudinal samples falls sharply with the

**Figure 3** Variation of tensile strength with LCP content for PC/ABS(M)/LCP/epoxy composites.**Figure 4** Variation of Young's modulus with LCP content for PC/ABS(M)/LCP/epoxy composites.

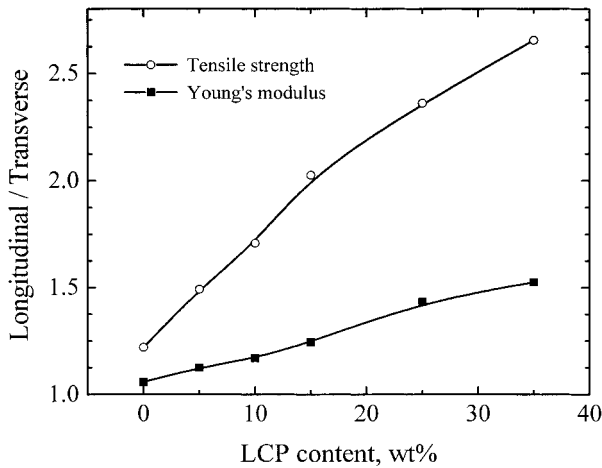


Figure 5 The ratios of tensile strength and Young's modulus of the longitudinal sample to those of the transverse sample vs LCP content for PC/ABS(M)/LCP/epoxy composites.

addition of only 5 wt % LCP. Thereafter, it decreases slowly with increasing LCP content. On the other hand, the incorporation of LCP into the PC/ABS blend does not result in a significant change of the strain at break for the transverse samples. Figure 7 shows the impact strength vs whisker content for PC/ABS(M)/LCP/Epoxy samples. This figure indicates that the impact strengths of these composites decrease with increasing LCP content up to 15 wt %; thereafter they increase slowly with increasing LCP content for both longitudinal and transverse specimens.

Figures 8(a) and (b) show the storage moduli vs. temperature for PC/ABS(M)/LCP/epoxy com-

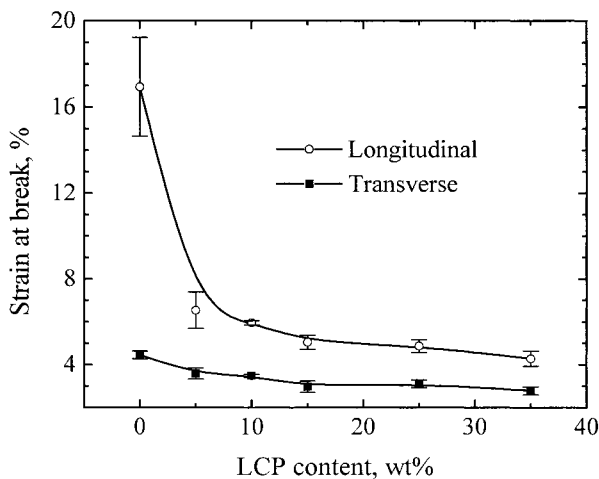


Figure 6 Variation of strain at break with LCP content for PC/ABS(M)/LCP/epoxy composites.

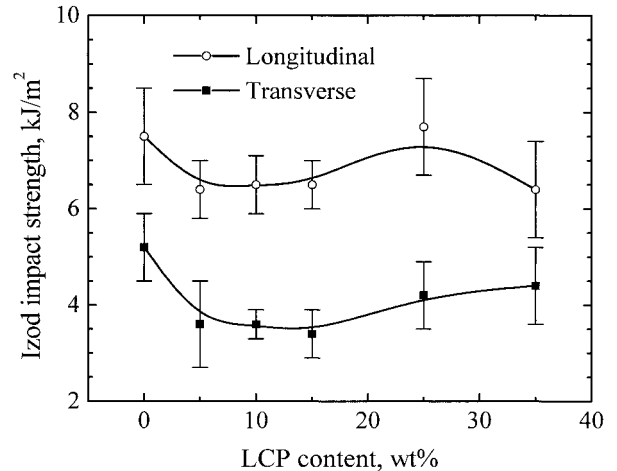


Figure 7 Variation of Izod impact strength with LCP content for PC/ABS(M)/LCP/epoxy composites.

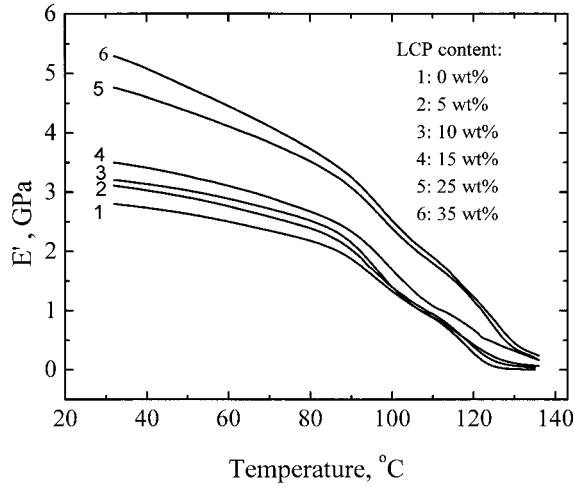
posites. From Figure 8(a), it can be seen that the storage modulus increases markedly with increasing LCP content from 30 to 135°C for the longitudinal samples. On the other hand, the storage modulus of transverse samples tends to decrease with increasing LCP content at temperatures up to 80°C; thereafter LCP addition has little effect on the storage modulus. All these results indicate that the mechanical strength of the longitudinal PC/ABS blends can be improved by adding LCP.

Thermal Properties

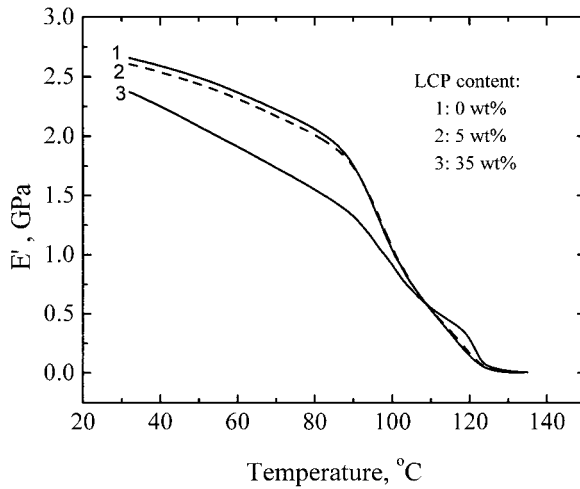
Figure 9 shows the weight loss vs temperature for the PC/ABS (60/40 by weight), LCP and PC/ABS(M)/LCP/epoxy specimens. As the LCP exhibits a higher decomposition temperature, the thermal stability of PC/ABS composite is improved considerably with increasing LCP content.

Morphology

Figure 10 shows a low magnification SEM fractograph of the injection molded PC/ABS(M)/25 wt % LCP/epoxy composite specimen. From this figure, it can be seen that this composite exhibits a typical skin-core structure. Figures 11 (a-d) are higher magnification views of the skin regions of the specimens containing different LCP contents. It is apparent that elongated LCP fibrils are well developed in the skin sections, even for the composite with a lower LCP content, i.e., 5 wt % [Fig.11(a)]. In this case, the tensile strength of the longitudinal sample improves dramatically with



(a)



(b)

Figure 8 Storage modulus spectra of (a) longitudinal and (b) transverse samples for PC/ABS(M)/LCP/epoxy composites.

increasing LCP (Fig. 3). On the other hand, the LCP phase tends to disperse as ellipsoids in the core sections of these samples. Figure 12 shows the SEM fractograph of the skin section of uncompatibilized PC/ABS/25% LCP composite. Comparing the morphology of the LCP fibrils of both compatibilized and uncompatibilized specimens, it is obvious that LCP fibrils formed in compatibilized composites are more finer, longer, and evenly distributed than those of uncompatibilized blend. Generally, two major factors are known to control the mechanical performance of the LCP/thermoplastic blends, i.e., the formation of the fibrillar LCP phase and the improvement of the

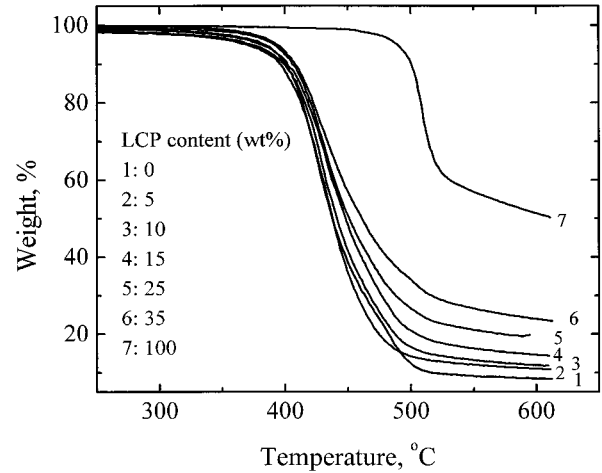


Figure 9 Weight loss vs temperature for LCP and PC/ABS(M)/LCP/epoxy composites.

interfacial adhesion. In this study, adhesion between the blend components (Fig. 2) and the formation of finer LCP fibrils (Fig. 11) give rise to a higher impact toughness and to a small improvement in tensile strength of MAP and epoxy compatibilized composite (PC/ABS/25wt%/epoxy/MAP), as shown in Table I.

CONCLUSIONS

Ternary PC/ABS/LCP blends were prepared by means of injection molding. Bisphenol type A epoxy resin was found to improve the compatibility between the blend components effectively. Tensile tests showed that the tensile strength of PC/ABS/LCP composite in the longitudinal direction increased markedly with increasing LCP content.

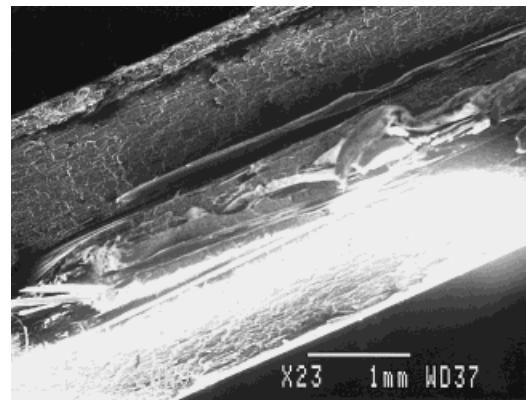


Figure 10 Low magnification SEM fractograph of PC/ABS(M)/25 wt%LCP/epoxy composite.

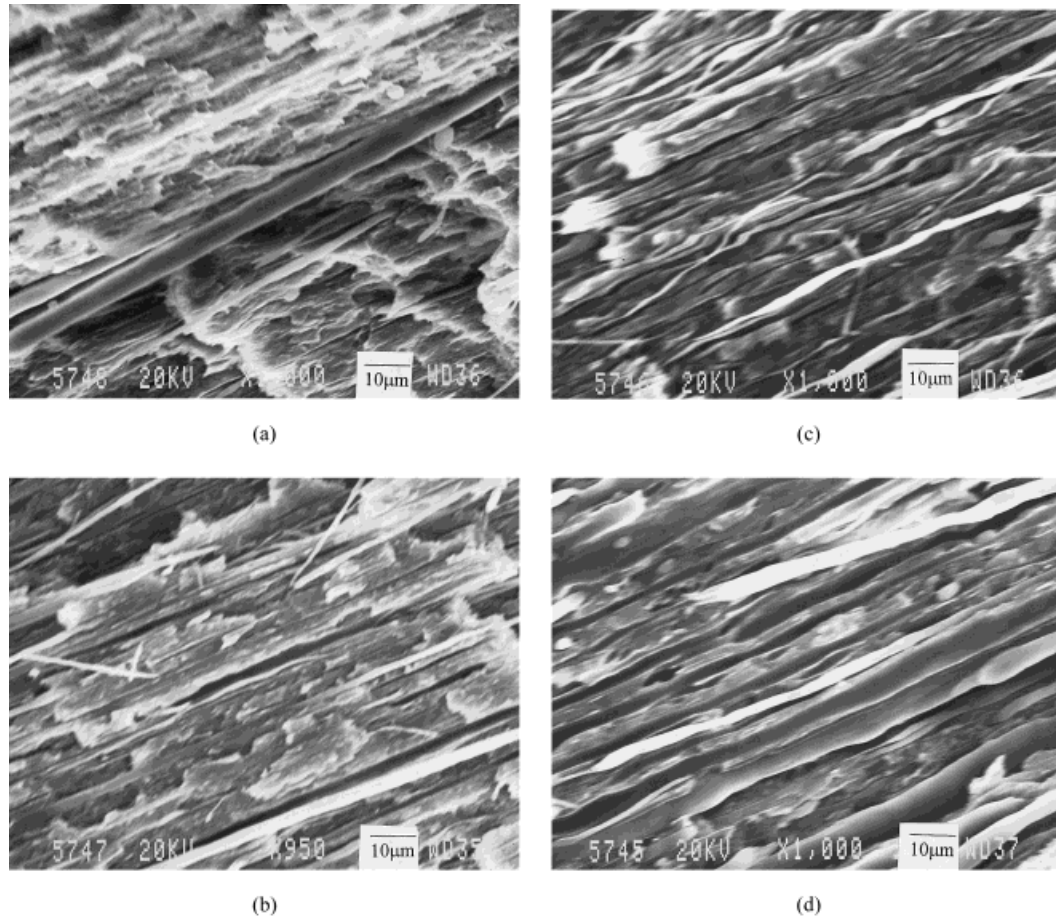


Figure 11 High magnification SEM fractographs of the skin section of PC/ABS(M)/LCP/epoxy composites: (a) 5 wt % LCP; (b) 10 wt % LCP, (c) 15 wt % LCP, and (d) 25 wt % LCP.

However, it decreased slowly with increasing LCP content in the transverse direction. The modulus of this composite in the longitudinal direction ap-

peared to increase considerably with increasing LCP content, whereas the incorporation of LCP into PC/ABS blends had little effect on the modulus in the transverse direction. The impact tests revealed that the Izod impact strength of the composite in both longitudinal and transverse directions decreased with the LCP content up to 15 wt % LCP; thereafter it increased slowly with increasing LCP content.

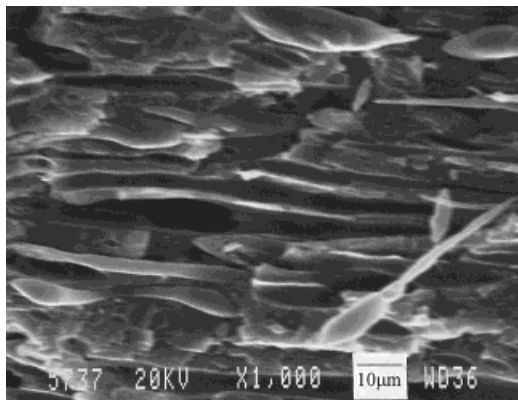


Figure 12 SEM fractographs of the skin section of uncompatibilized PC/ABS/25 wt % LCP composite.

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