The processing of ternary LCP/LCP/thermoplastic blends*

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The feasibility of introducing a second thermotropic liquid crystalline polymer (TLCP) as a compatibilizer, or coupling agent, in order to improve the adhesion and dispersion between components of incompatible TLCP/thermoplastic blends was investigated. Two TLCP blends of a wholly aromatic copolyester (K161) with poly(ethylene terephthalate) (PET) and polycarbonate (PC) were prepared by a two-step melt-blending process, using an aliphatic containing TLCP, PET/60% poly(hydroxybenzoate) (PHB60), as the coupling phase. Preliminary morphological evidence indicated that the K161 reinforcing phase in the ternary systems exhibited improved adhesion and dispersion in a much finer scale than in binary blends of K161/PET and K161/PC prepared under the same processing conditions. It was found that the adhesion between the reinforcing K161 and the matrix PET phases depended on both the concentration of the coupling PET/PHB60 phase and the relative composition of the two TLCPs. The improved adhesion between K161 and PC matrix with the inclusion of PET/PHB60 phase was further delineated by comparing the phase morphology and mechanical properties of the ternary blends after a cold-drawing process with those of the binary systems. While the binary systems remained practically unaffected by the cold-drawing process, the ternary blends exhibited a well aligned and elongated K161 fibrillar phase with a diameter of around 1 μm or less. An increase of both initial modulus and breaking strength suggests that the enhanced adhesion between K161 and PC facilitates a better stress transfer for the deformation of the dispersed K161 particles into oriented, ellipsoidal domains. Other characterizations, including both differential scanning calorimetry and dynamic mechanical analysis, were conducted to elucidate the nature of the interactions between the components.

(Keywords: thermotropic liquid crystalline polymers; polymer blends; compatibilizing agent; coupling agent; processing)

INTRODUCTION

Thermotropic liquid crystalline polymers (TLCPs), featuring mesomorphic melts (i.e. anisotropic melts) within a defined temperature range, have attracted considerable attention in recent years. For the most part, the interests in TLCPs lie in their excellent balance of mechanical performance and melt processability in the liquid crystalline state. When blended with a conventional thermoplastic resin, the melt-processable TLCP phase not only can serve as a processing aid by lowering the melt viscosity of the material, but is also a prime candidate for improving the mechanical performance of the host thermoplastic matrix by forming fibrous reinforcements *in situ*. A considerably number of studies have been conducted to elucidate the rheology and thermodynamics of binary TLCP/thermoplastic blends as well as the processing variables controlling the morphology and the mechanical properties[1–10]. It is realized that the mechanism of forming an elongated TLCP fibrous structure *in situ* is controlled by factors such as the relative composition, the interfacial tension, the viscosity ratio and the processing conditions[11]. Moreover, because of their thermodynamic incompatibility, poor adhesion between TLCPs and thermoplastic matrices has been a limiting factor in the development of TLCP reinforced composite systems.

Compared to the vast number of studies on TLCP/thermoplastic blends, limited effort has been directed towards blends containing two TLCPs. Jin *et al.*[12] studied the miscibility of two nematic TLCPs and concluded that in the same mesomorphic state the two TLCPs were miscible and would phase separate in the solid state. Miscibility between two TLCPs has also been reported by Kiss[13] for a binary TLCP blend of a poly(ester–amide) and a copolyester containing hydroxybenzoic acid and hydroxynaphthoic acid. De Meuse and Jaffe[14–16] conducted a series of detailed studies on TLCP/TLCP blends and concluded that the miscibility of two TLCPs depended on the composition of the copolyester, and both the entropic and enthalpic portions of the free energy of mixing.

Approaches that have been used to improve adhesion between *in situ* formed TLCP fibrils and thermoplastic matrices include modification of the geometry of the TLCP fibres to provide mechanical locking to the matrix[17] and chemical grafting of flexible polymers to the TLCP fibre surface[18]. In a previous study[19], the idea of using a partially compatible and/or reactive TLCP as a 'coupling agent' was introduced in order to improve the adhesion between components in normally incompatible TLCP/thermoplastic blends, in much the same fashion as a compatibilizer in other polymer blend

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systems. In that particular system, the chemical interaction between the reinforcing TLCP (K161 phase) and the coupling TLCP (poly(ethylene terephthalate)/60% poly(hydroxybenzoate), PET/PHB60) resulted in a two-phase morphology with the K161-rich phase dispersed in and strongly bonded to a fibrillar PET/PHB60-rich matrix phase. The adhesion observed most likely resulted from a molecular exchange reaction initiated by the thermally unstable PET/PHB60 phase under the blending conditions employed. The potential compatibility of PET/PHB60 with some engineering thermoplastics such as PET[20], polycarbonate (PC)[8,10] and poly(hexamethylene terephthalate) (PHMT)[21] offers the possibility of obtaining a ternary blend system using PET/PHB60 as a compatibilizing agent. In the case of blending PET/PHB60 with PET or PC, the compatibility is thought to originate from partial miscibility of the PET-rich portion of PET/PHB60 with the thermoplastic[8,10,11,20], whereas transesterification is responsible for the compatibilization of (PET/PHB60)/PHMT blends[21,22].

An inclusion of a second TLCP in an incompatible TLCP/thermoplastic blend not only leads potentially to a ternary system with improved adhesion due to physical or chemical compatibilization, but also provides the advantage of enhanced processability. According to Cogswell[23], melt blending of a TLCP with a thermoplastic requires the processing temperature range of the two components to overlap to an extent of at least 5°C or more so that the thermoplastic is in its molten state and the TLCP is in the anisotropic melt state. Attempts to melt blend a TLCP with a thermally distinctive thermoplastic usually result in a decomposition of the lower melting constituent and/or poor dispersion[23]. This requirement certainly limits the choice of possible TLCP/thermoplastic pairs. For example, the copolyester containing *p*-hydroxybenzoic acid and biphenol terephthalate (BPT) (trade name Xydar, Dart Kraft Co.) has been demonstrated to have outstanding fibre properties and thermal stability[24,25]. However, this copolymer does not melt until about 400°C, which makes it difficult to blend with traditional engineering plastics without the occurrence of thermal degradation of the thermoplastics. By blending the thermally stable, reinforcing TLCP with a second, lower-melting TLCP coupling phase, the processing temperature window can conceivably be tailored to an appropriate range.

It is the purpose of this continuing study to examine the feasibility of using a second TLCP as a compatibilizing, or coupling, agent to improve the adhesion between incompatible TLCP/thermoplastic phases. Two TLCP ternary systems were prepared using PET and PC as the matrices. The effects of the coupling phase on the morphology and mechanical properties of the ternary blends were investigated.

EXPERIMENTAL

The TLCP ternary blends were prepared through a two-step melt-blending process. The TLCP/TLCP binary blends containing K161 (Bayer A.G., KU-9211) and PET/PHB60 (Tennessee Eastman) of various proportions were first melt-blended following the procedures described in a previous publication[19]. The as-prepared TLCP/TLCP blends were then ground into powder for subsequent processing. Two thermoplastic matrices chosen were PET (Kodak Tennessee Eastman Co., formula number PM-9663, batch number C6011) and PC (G.E., Lexan 181). Before blending, both thermoplastic resins were dried overnight under vacuum at around 100°C. The ternary blends were then prepared by melt-blending the binary TLCP/TLCP mixture with PET and PC respectively in a Brabender at 280°C and 40 rev min^{-1} for only 5 min to avoid possible thermal or mechanical degradation of the thermoplastics. These ternary blends were hot-pressed into sheets using a Wabash press at 270°C for PET and 200°C for PC, followed by water cooling under pressure at around 9.1–13.6 t.

The morphology of fracture surfaces of these blends was studied using scanning electron microscopy (SEM). The fracture surface was obtained by breaking the hot-pressed sheets in liquid nitrogen. Thermal characterizations were conducted by differential scanning calorimetry (d.s.c.) using a Du Pont DSC-910 at a heating rate of 10°C min^{-1}. Dynamic mechanical thermal analysis (d.m.t.a.) of the blends was carried out with a Polymer Laboratories instrument using the tensile mode at a frequency of 1 Hz. The mechanical properties of the blends were measured following ASTM D-638. Dog-bone specimens were prepared by hot-pressing the as-mixed blends with dimensions following type V sample specifications. The stress–strain response was measured at room temperature using an Instron model 1011 testing machine at a cross-head speed of 1.27 mm min^{-1}. The initial modulus, yield strength, fracture strength and strain at break were calculated both before and after cold-drawing of the specimens.

RESULTS AND DISCUSSION

Figure 1 is an SEM micrograph of the ternary blend containing 21 wt% of K161 as the reinforcing phase, 70 wt% of PET as the matrix phase, and 9% of PET/PHB60 as the coupling agent. As can be seen, K161 spherical domains with dimensions in the range of 1 μm or less are well dispersed in and bonded to the PET matrix. A close-up of the K161/(PET/PHB60)/PET interface is shown in *Figure 2* indicating very good adhesion between all components. For comparison, *Figure 3* is an SEM micrograph of a K161/PET blend (21/70 weight ratio) without the coupling PET/PHB60

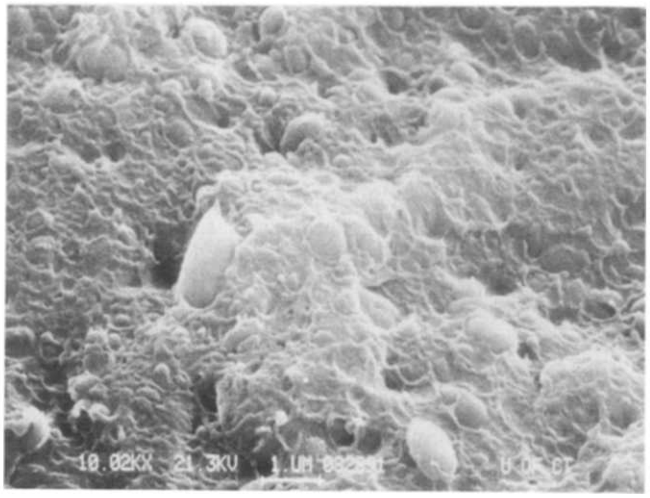

Figure 1 SEM micrograph of the as-prepared K161/(PET/PHB60)/PET ternary blend

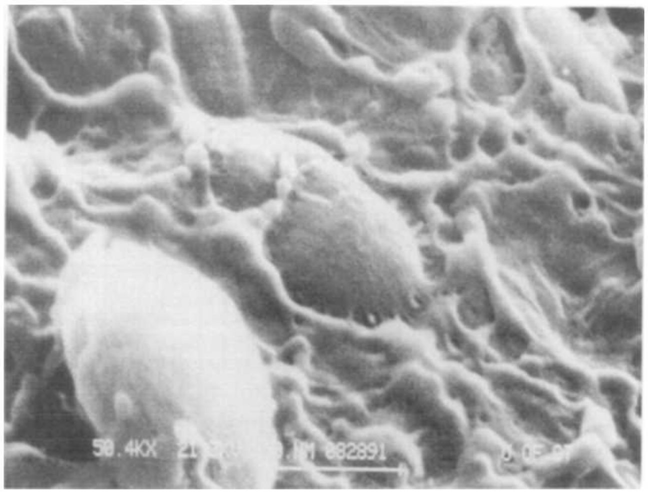

Figure 2 Close-up of the interface in the as-prepared K161/PET(PHB60)/PET ternary blend

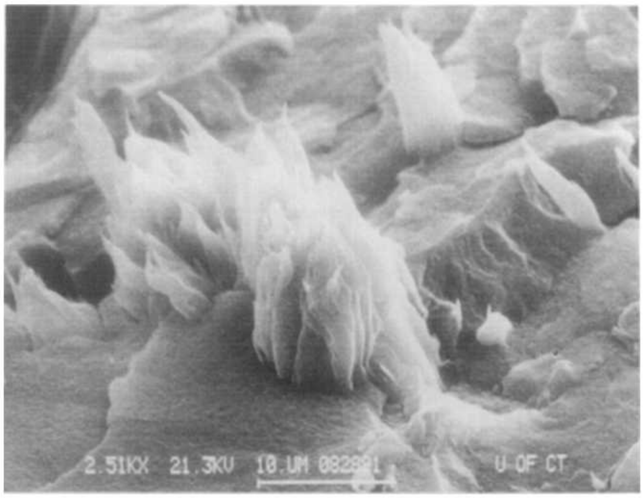

Figure 3 SEM micrograph of the as-prepared K161/PET binary blend

material, prepared under exactly the same mixing conditions as the ternary systems. It appears that without the presence of PET/PHB60 in the system, the dispersion of K161 in PET is poorer, resulting in dispersed K161 domains in the order of 1–10 μm in diameter. Also noted is the poor adhesion between K161 and PET, as expected, since they have inherently different molecular structures.

The tan δ response of the ternary and the binary blends using PET as the matrix material as a function of temperature are compared in *Figures 4* and *5*. As can be seen, the ternary blend (*Figure 4*) exhibits a shifted tan δ peak at around 122°C, intermediate between those of the constituents, along with a shoulder located at around 105°C. The binary K161/PET mixture, on the other hand, exhibits two distinct damping peaks (*Figure 5*), which is characteristic of an immiscible two-phase system[26]. Since the two blends contain similar amounts of K161 and were prepared under the same mixing conditions, the difference in the dynamic relaxation spectra can be attributed to the presence of the PET/PHB60 phase in the ternary systems. Based on the d.m.t.a. results alone, however, it is difficult to determine the role of PET/PHB60 because the per cent crystallinity of the PET could also vary from system to system[20]. The per cent crystallinity of the PET, measured by d.s.c., was 30, 39, 43 and 31 for neat PET, PET in a blend with K161, PET in PET/PHB60 and PET in the ternary blend, respectively. Thus, it is difficult to separate the effect of compatibilization from that of crystallization. Nonetheless, the difference in the dynamic relaxation spectra of the two blends is consistent with the morphological results which suggest that the inclusion of PET/PHB60 in the ternary blends improves the adhesion and dispersion of K161 in PET matrix.

The d.s.c. thermograms of the ternary blend, pure PET and K161/(PET/PHB60) (21/9) are compared in *Figure 6*. As can be seen, the ternary blend exhibits a lower crystallization temperature relative to that of the pure PET, along with a glass transition temperature intermediate between the individual components. In comparison, the thermogram of a K161/PET blend (*Figure 7*) demonstrates a similar lowering of the crystallization peak, while the glass transition remains the same as that of the pure PET. The lowering of the crystallization temperature has been attributed to the presence of the TLCP phase which serves as nucleating agent to enhance

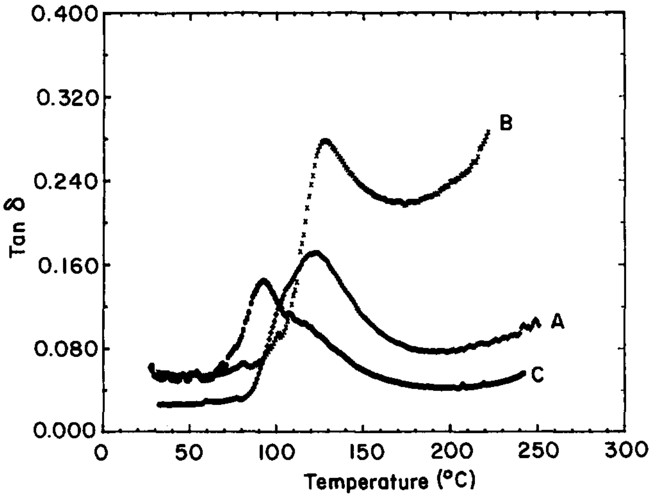

Figure 4 Comparison of the dynamic spectra between K161/(PET/PHB60)/PET ternary blend and its constituents. A, K161/(PET/PHB60)/PET (21/9/70); B, K161/(PET/PHB60) (21/9); C, PET

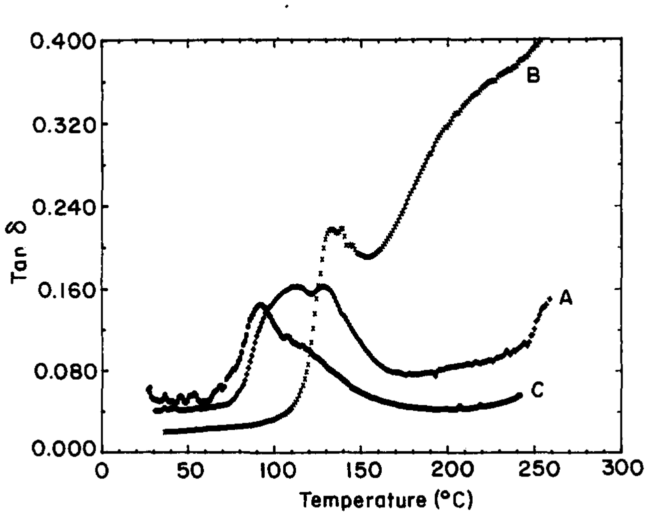

Figure 5 Comparison of the dynamic spectra between K161/PET binary blend and its constituents. A, K161/PET (21/70); B, K161; C, PET

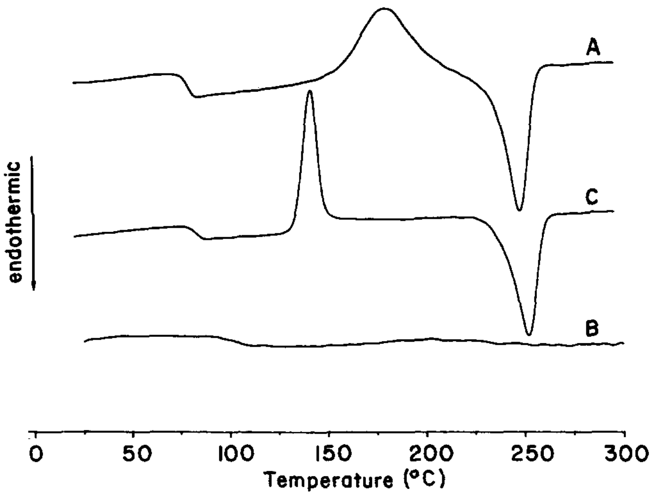

Figure 6 D.s.c. thermograms: A, PET; B, K161/(PET/PHB60) (21/9), C, the resultant ternary blend

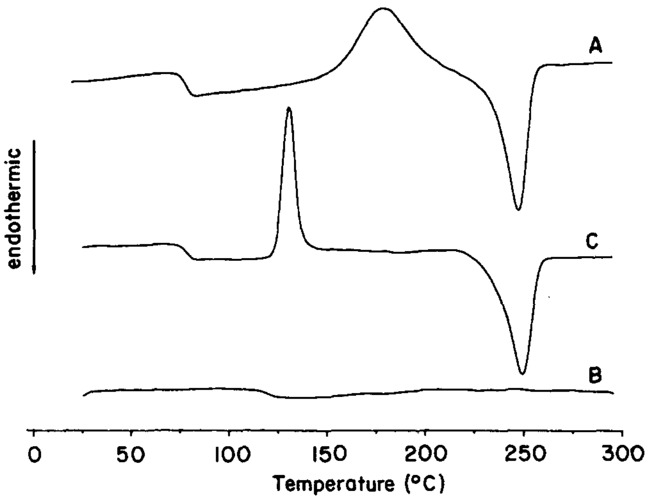

Figure 7 D.s.c. thermograms: A, PET; B, K161; C, K161/PET binary blend

the crystallization of the PET[20]. The intermediate glass transition observed in the ternary blend suggests an improved compatibility compared to the binary TLCP/thermoplastic blend. It has been reported[27] that the transesterification reaction of PET blended with polyarylate requires an induction time of about 15 min at 280°C. Therefore, under the blending conditions employed, it seems that the improved compatibility observed in the ternary system might be due to the partial miscibility between the PET matrix and the PET/PHB60 coupling phase, although no conclusive evidence is available for the present.

Morphological evidence along with d.m.t.a. and d.s.c. results for the ternary TLCP blends, using PET as the matrix, support the concept of introducing a second TLCP as the compatibilizing phase. The poor adhesion and dispersion of K161 in PET with the absence of PET/PHB60 are associated with the magnitude of the interfacial tension as well as their distinct thermal characteristics. The binary blend also presents a typical example of a TLCP/thermoplastic blend with a very narrow processing window between the melting point of the PET matrix (250°C) and the softening temperature of K161 (280°C). Blending at a temperature above 280°C usually causes PET to suffer thermal or thermo-oxidative degradation[28,29], while any processing temperatures below 280°C result in poor mixing due to the relatively high viscosity of K161. With the introduction of PET/PHB60 as the intermediate phase, which is potentially miscible and/or chemically interactive with both the reinforcing K161 phase and the PET matrix, the processing of the two thermally distinctive materials becomes possible and results in a ternary system exhibiting improved adhesion and a much finer scale of dispersion of the TLCP phase. When K161/(PET/PHB60) blends of different proportions were mixed with PET, it was found that the adhesion between the TLCP blend and the PET matrix was a function of both the absolute content of PET/PHB60 and the relative amount of the two TLCPs in the ternary system. *Table 1* summarizes the goodness of adhesion in the ternary blends of different compositions, based on qualitative observations of SEM micrographs.

The feasibility of using PET/PHB60 as a compatibilizing, or coupling, agent was also supported by examining the phase morphology of the ternary blends using PC as the matrix material. *Figure 8* shows an SEM micrograph of a ternary blend containing 21 wt% of K161, 9 wt% of PET/PHB60 and 70 wt% of PC. Similar to PET ternary blends, the reinforcing K161 phases are well adhered to and dispersed in the PC matrix with dimensions of around 3 μm or less in diameter. *Figure 9* is an example of the fracture surface of a K161/PC binary blend (21/70 weight ratio) prepared under the same processing conditions. In contrast to that of the ternary systems, the

Table 1 Adhesion between K161/(PET/PHB60) blends and PET

TLCP/TLCP fibre blend K161/(PET/PHB60)	Fibre blend (wt%)	PET/PHB60 in TLCPs/PET composite (wt%)	PET (wt%)	Adhesion between phases
100/0	30	0	70	Poor
90/10	30	3	70	Good
70/30	30	9	70	Good
50/50	30	15	70	Good
30/70	30	21	70	Poor
30/70	10	7	90	Poor
10/90	10	9	90	Poor

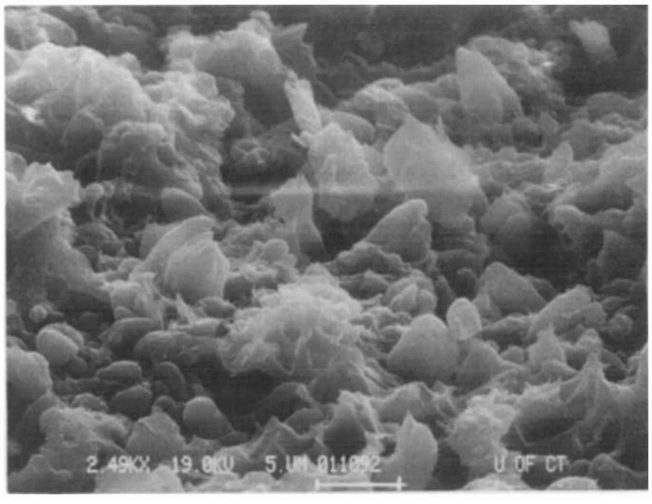

Figure 8 SEM micrograph of the as-prepared K161/(PET/PHB60)/PC ternary blend

fracture surface demonstrates considerable matrix dilation, which is a phenomenon usually associated with systems of poor bonding between components.

As to the nature of the adhesion observed in the ternary blends, thermal analyses, including both d.s.c. and d.m.t.a., indicated that neither a chemical reaction nor an enhanced (partial) miscibility seems to have occurred between the K161/(PET/PHB60) mixture and PC. The transitions in the ternary blends can be superposed over those of the neat constituents. Therefore, it is speculated that the improved adhesion observed in the electron micrographs of the TLCPs/PC ternary blends is most likely attributed to an improved wetting between PC matrix and the dispersed TLCP/TLCP surface due to the rheological properties of the PET/PHB60 intermediate layer and the interfacial tension of the resulting interface. More detailed characterizations are in progress.

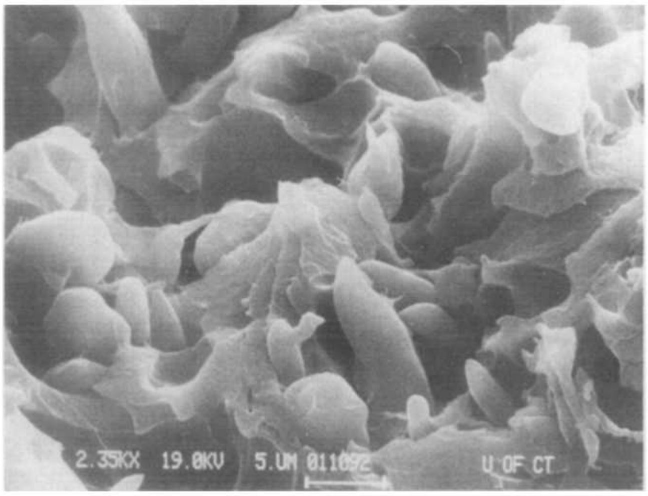

Figure 9 SEM micrograph of the as-prepared K161/PC binary blend

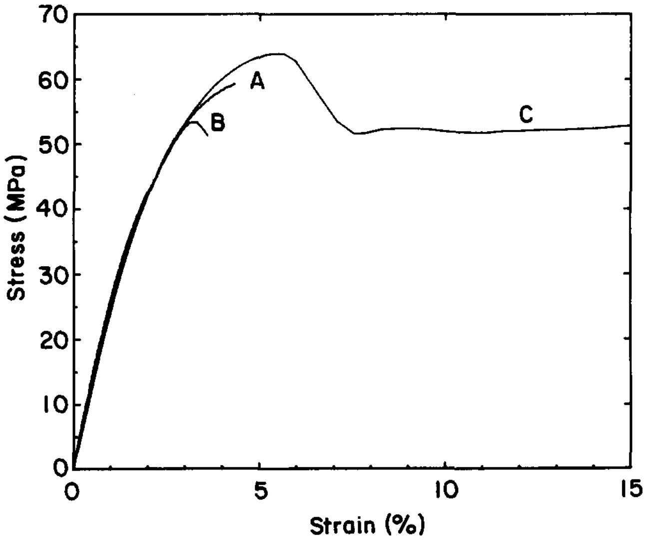

Figure 10 Stress–strain curves of as-prepared ternary (A) and binary (B) blends measured at room temperature in comparison with that of PC (C)

Figure 10 shows a typical stress–strain response of the ternary blends compared with those of K161/PC binary blends and pure PC measured at room temperature. The calculated mechanical properties are listed in *Table 2.* As shown, the ternary blends exhibit brittle behaviour with the ultimate strength and elongation at break somewhat higher than those of the binary systems. It can also be seen that neither blend attains the mechanical properties of the host PC matrix. This result, along with the phase morphology of the as-prepared blends (*Figures 8* and *9*), leads to the conclusion that in order to strengthen and stiffen the composite, the embedded TLCP phase has to be elongated into fibres and aligned in the loading direction. The initial moduli of the K161 and PET/PHB60 in their non-drawn state are around 3–5 GPa, but can be increased to 30–60 GPa by hot-drawing[30].

The formation of a fibrous TLCP phase *in situ* is controlled by factors such as the viscosity ratio, interfacial tension, relative composition and the processing conditions[11]. The general approach reported in the literature has been to extrude or draw the blends in their molten state. In this work tensile samples of the ternary blends were cold-drawn at reduced rates and elevated temperatures, allowing stress transfer from the PC matrix to the dispersed phases to elongate and align the TLCP particles *in situ*. The conditions for preparing cold-drawn samples are obtained by examining the stress–strain curves of the as-prepared blends tested at various temperatures and strain rates. *Figure 11* illustrates the stress–strain response of the ternary blends measured at different temperatures and a cross-head speed of 0.508 mm min^{-1}. By increasing the testing temperature, the ternary blends undergo a brittle–ductile transition, resulting in a strain hardening prior to ductile failure. At around 100°C, a stable neck is formed throughout the whole gauge length, enabling elongation and orientation of the dispersed TLCP particles. The draw ratio is defined as the ratio of cross-sectional area in the gauge of the dog-bone samples before and after the cold-drawing process.

Figure 12 illustrates the mechanical properties at 25°C and three different draw ratios of the ternary blends after cold-drawing at 100°C. The stress–strain curves were measured at a cross-head speed of 1.27 mm min^{-1} so that a direct comparison of the mechanical behaviour can be made with that of the non-drawn ternary blends. The tensile properties of the blends are summarized in *Table 3* and compared to those of neat, cold-drawn PC. It can be seen that the cold-drawn ternary blends exhibit a substantial increase of the tensile properties compared to those of the as-prepared blends without drawing. When compared with cold-drawn PC, the tensile modulus

Table 2 Mechanical properties of as-prepared ternary and binary blends measured at room temperature in comparison with those of PC

Materials	Number of test specimens	Yield stress (MPa)	Yield strain (%)	Tensile strength (MPa)	Strain at break (%)	Young's modulus (GPa)
PC	10	63.7±2.1	5.5±0.2	60.4±5.8	127.7±17.4	2.4±0.2
K161/(PET/PHB60)/PC ternary blend (21/9/70)	8	–	–	59.6±1.6	4.4±0.8	2.6±0.1
K161/PC binary blend (21/70)	7	52.5±2.3	3.1±0.2	49.4±2.8	3.4±0.7	2.6±0.1

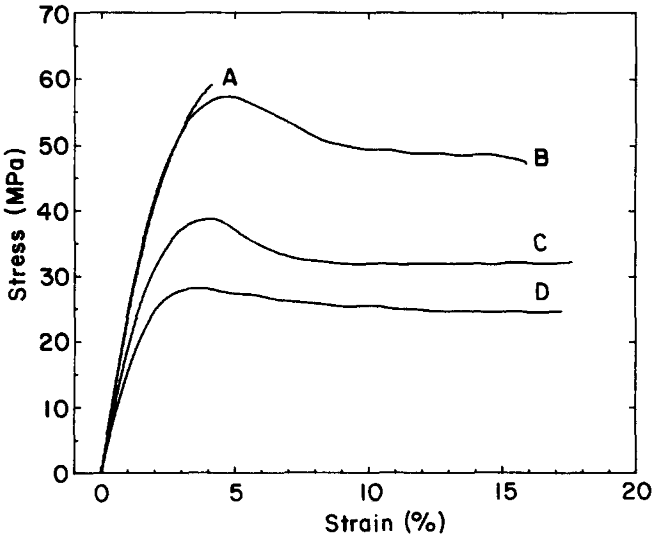

Figure 11 Stress–strain curves of as-prepared ternary blends at various temperatures: A, 25°C; B, 50°C; C, 75°C; D, 100°C. Cross-head speed 0.508 mm min^{-1}

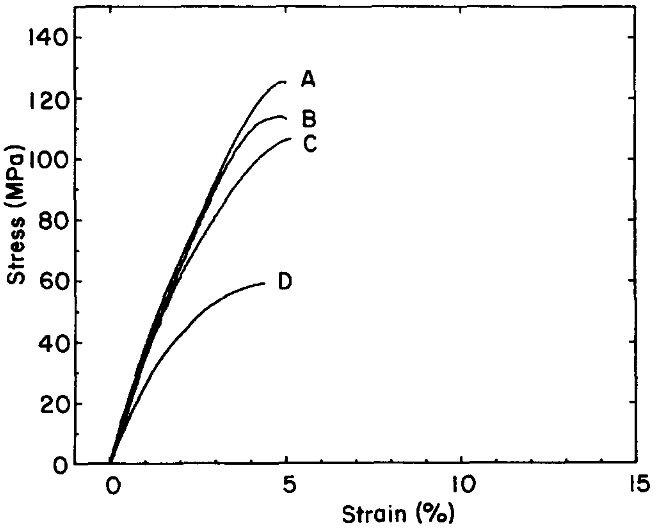

Figure 12 Stress–strain curves of a K161/(PET/PHB60)/PC blend measured at 25°C. A–C, after cold-drawing at 100°C to draw ratio: A, 1.93; B, 1.61; C, 1.57. D, before cold-drawing

Table 3 Mechanical properties of cold-drawn ternary and binary blends measured at room temperature in comparison with those of PC after cold-drawing

Materials	Cold-draw ratio	Ultimate strength (MPa)	Strain at break (%)	Young's modulus (GPa)
K161/(PET/PHB60)/PC ternary blends (21/9/70)	1.93	124.8	5.0	4.0
	1.61	113.9	5.0	3.6
	1.57	106.6	5.1	3.5
K161/PC binary blends (21/70)	1.65	103.2	4.2	3.3
PC	1.77	102.0[a]	5.2[b]	3.2

[a] Yield stress of PC after cold-drawing

[b] Yield strain of PC after cold-drawing

Note: data are obtained from single measurements on each of the cold-drawn samples

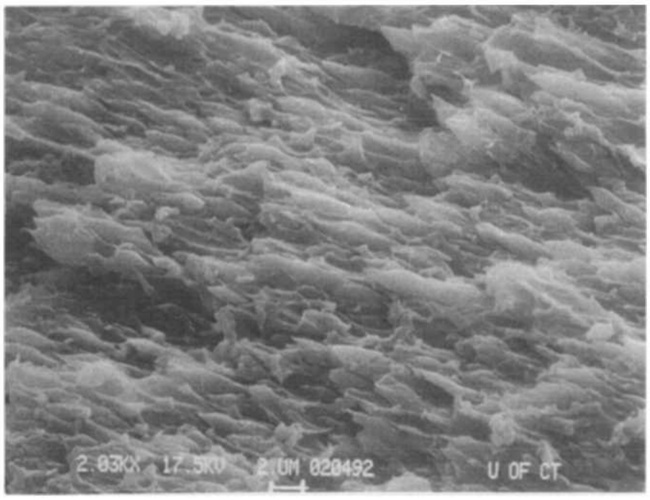

Figure 13 SEM micrograph of longitudinal fracture surface of K161/(PET/PHB60)/PC ternary blend after cold-drawing

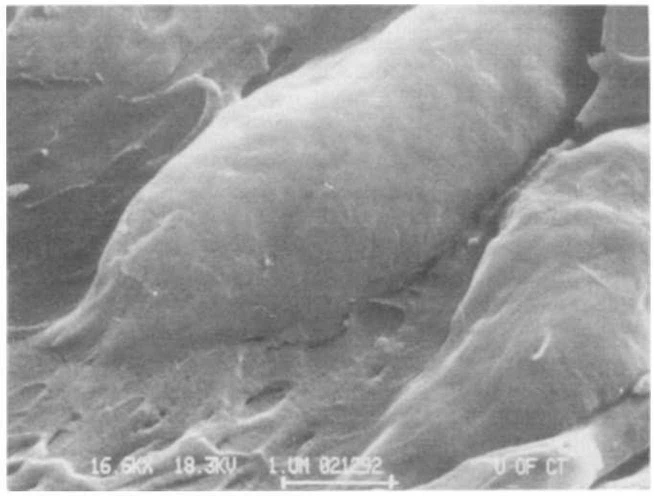

Figure 14 Close-up of the interface between K161 and PC in the cold-drawn ternary blend

increases 10–25% and the ultimate strength increases 5–23%, depending on the draw ratio. The improvement in the mechanical properties can be explained by examining the corresponding phase morphologies. *Figure 13* shows an electron micrograph of the longitudinal fracture surface obtained in liquid nitrogen of a ternary blend cold-drawn in the range of 1.5–2. The elongated K161 domains have diameters in the order of 1–2 μm or less and are well aligned in the drawing direction. A close-up of the interface between K161 and PC is shown in *Figure 14*, demonstrating that strong bonding exists between all constituents. It is believed that the good bonding of components facilitates the transfer of the applied stress to the dispersed K161, resulting in a morphology of moderately elongated and well oriented K161 fibrils. *Figure 15* shows an SEM micrograph of a transverse fracture surface of the ternary blend where the TLCP fibres seem to be well bonded to the PC matrix. The maximum draw ratio reported was about 2, but appropriate optimization of the drawing conditions should enable draw ratios of the order of at least 5–10. The binary K161/PC blends were also cold-drawn under the same conditions as the ternary systems. However, complete strain-hardening was preceded by premature

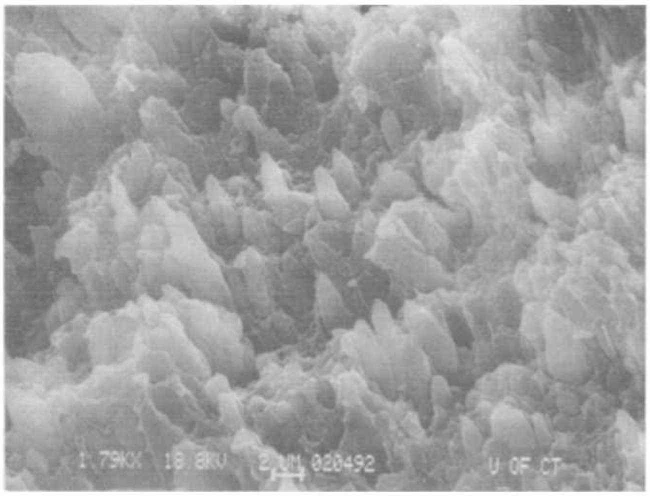

Figure 15 SEM micrograph of transverse fracture surface of K161/(PET/PHB60)/PC ternary blend after cold-drawing

Figure 16 SEM micrograph of transverse fracture surface of K161/PC binary blend after cold-drawing

failure of the material, preventing a uniform necking in the gauge. *Figure 16* shows a transverse fracture surface of K161/PC binary blend cold drawn at 100°C and cross-head speed of 0.508 mm min^{-1}. As can be seen, without the inclusion of the PET/PHB60 coupling phase the cold-drawing process does not impose substantial deformation or alignment of the dispersed K161 phase. There is also considerable dilation of the matrix around the K161 particles, indicating poor adhesion between the two components. An estimate of the mechanical properties of cold-drawn binary blends of draw ratio around 1.65 indicates that the tensile properties are similar to those of the cold-drawn PC with an initial modulus around 3.3 GPa and an ultimate strength around 103.2 MPa.

CONCLUSIONS

The work presented here supports the concept of employing a second TLCP to serve as a compatibilizing, or coupling, agent to improve the adhesion and dispersion of incompatible TLCP/thermoplastic blends. Morphological evidence demonstrated that a highly aromatic TLCP can be finely dispersed and well bonded to an inherently incompatible PET matrix through the presence of a second TLCP as a compatibilizing component. In the systems studied, the adhesion obtained was probably associated with either partial miscibility or chemical reaction of the intermediate layer with the thermoplastic matrix. The effectiveness of the coupling depended on both the absolute content of the compatibilizing phase and the relative composition of the two TLCPs. While binary blends of the aromatic fibre and PC were relatively unaffected by a cold-drawing process, cold-drawn ternary blends exhibited elongated and well aligned TLCP fibres and improved tensile properties, suggesting that good adhesion facilitated the transfer of applied stress from the PC matrix to the TLCP, enabling *in situ* fibre formation.

It is envisioned that this study can eventually lead to the development of *in situ* coupling between reinforcing thermotropic fibres and incompatible thermoplastic matrices using well controlled reaction mechanisms such that during melt processing an intermediate compatibilizing phase could be formed *in situ*. A successful preparation of *in situ* compatibilized ternary blend systems would seemingly stimulate more interest in the development of TLCP/thermoplastic blends.

REFERENCES

1 Weiss, R. A., Huh, W. and Nicolais, L. in 'High Modulus Polymers' (Eds A. E. Zachariades and R. S. Porter), Marcel Dekker, New York, 1988, Ch. 5
2 Brostow, W. *Polymer* 1990, **31**, 979
3 Siegmann, A., Dagan, A. and Kenig, S. *Polymer* 1985, **26**, 1325
4 Chung, T.-S. *ANTEC Tech. Paper* 1987, 1404
5 Swaminathan, S. and Isayev, A. I. *Proc. ACS Div. Polym. Mater.* 1987, **57**, 330
6 Kiss, G. *Polym. Eng. Sci.* 1987, **27**(6), 410
7 Blizard, K. G. and Baird, D. G. *Polym. Eng. Sci.* 1987, **27**(9), 653
8 Zhuang, P., Kyu, T. and White, J. L. *Polym. Eng. Sci.* 1988, **28**(17), 1095
9 Kimura, M. and Porter, R. S. *J. Polym. Sci., Polym. Phys. Edn* 1984, **22**, 1697
10 Friedrich, K., Hess, M. and Kosfeld, R. *Macromol. Chem. Macromol. Symp.* 1988, **16**, 251
11 Sukhadia, A. M., Done, D. and Baird, D. G. *Polym. Eng. Sci.* 1990, **30**(9), 519
12 Jin, J.-I., Choi, E.-J. and Lee, K.-Y. *Polym. J.* 1986, **18**(1), 99
13 Kiss, G. D. US Patent, 4 567 227, 1986
14 De Meuse, M. T. and Jaffe, M. *Mol. Cryst. Liq. Cryst. Inc. Nonlin. Opt.* 1988, **157**, 535
15 De Meuse, M. T. and Jaffe, M. in 'Liquid-Crystalline Polymers' (Eds R. A. Weiss and C. K. Ober) ACS Symposium Series 435, American Chemical Society, Washington, DC, 1990, Ch. 30
16 De Meuse, M. T. and Jaffe, M. *ANTEC Tech. Papers* 1991, 905
17 Shaw, M. T. and Rueda, G. *Polym. Prepr.* 1989, **30**(2), 538
18 Datta, A., De Souza, J. P., Sukhadia, A. M. and Baird, D. G. *ANTEC Tech. Papers* 1991, 913
19 Lee, W. and DiBenedetto, A. T. *Polym. Eng. Sci.* 1992, **32**(6), 400
20 Joseph, E. G., Wilkes, G. L. and Baird, D. G. in 'Polymeric Liquid Crystals' (Ed. A. Blumstein), Plenum, New York, 1985, p. 197
21 Croteau, J.-F. and Laivins, G. V. *J. Appl. Polym. Sci.* 1990, **39**, 2377
22 Laivins, G. V. *Macromolecules* 1989, **22**, 3974
23 Cogswell, F. N. US Patent, 4433083, 1984
24 Economy, J. *J. Macromol. Sci.-Chem.* 1984, **A21**(13&14), 1705
25 Economy, J. *Mol. Cryst. Liq. Cryst.* 1989, **169**, 1
26 Murayama, T. 'Dynamic Mechanical Analysis of Polymeric Material', Materials Science Monographs, 1, Elsevier Scientific, New York, 1978, Ch. 3
27 Kimura, M., Salee, G. and Porter, R. S. *J. Appl. Polym. Sci.* 1984, **29**, 1629
28 Yoda, K., Tsuboi, A., Wada, M. and Yamadera, R. *J. Appl. Polym. Sci.* 1970, **14**, 2357
29 Nearly, D. L. and Adams, L. J. *J. Polym. Sci. A-1* 1971, **9**, 2063
30 Lee, W., DiBenedetto, A. T., Gromek, J. M., Nobile, M. R. and Acierno, D. *Polym. Eng. Sci.* in press