

# Solid State Structure and Mechanical Properties of Melt Mixed Poly(trimethylene terephthalate)/Polycarbonate blends

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**ABSTRACT:** Poly(trimethylene terephthalate) (PTT)/poly(carbonate of bisphenol A) (PC) blends were obtained in the melt state by direct injection molding and also by extrusion followed by injection molding. The blends rich in PTT were monophasic, while the blends rich in PC were biphasic with the two components of the blends present in both phases. Both the monophasic and biphasic blends were partially miscibilized, and also partially reacted, as observed by FTIR. The extent of the reaction was greater in previously mixed blends. The observed synergism in the modulus of elasticity was attributed to the

increased orientation of the blend components upon blending. Although decreases in elongation at break were observed and attributed to degradation of PTT, the blends were clearly ductile and compatible. This was a consequence of either their monophasic structure, or of the presence of the two components in the two phases of the blends. © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 108: 3828–3835, 2008

**Key words:** blends; mechanical properties; melt; polycarbonates; polyesters

## INTRODUCTION

Polymer blending continues to be one of the most attractive ways to develop new polymeric materials.<sup>1</sup> This is due to the versatility that is offered mainly by (i) the chemical structure of the components that may lead to interactions and reactions between the components of the blends, which are different in nature and strength, and (ii) the nature, either thermoplastic or rubbery of the components of the blends. These variables give rise to either miscible or biphasic blends and in the latter case to (i) different compositions of the dispersed phases, (ii) different interphase conditions, and (iii) different dispersed phase morphologies, that consequently produce both immiscible and biphasic blends with a wide range of mechanical properties, among others.

When a new commercial polymer is produced, its blends become rapidly the object of research interest both from a basic and an applied point of view. Poly(trimethylene terephthalate) (PTT) is a polymer that belongs to the aliphatic–aromatic polyester family, of which poly(ethylene terephthalate) (PET) and poly(butylene terephthalate) (PBT) are the most used

components. PTT shows the chemical resistance of polyesters, and the resilience and elastic recovery typical of the polyamide 66.

The blends of PTT with poly(carbonate of bisphenol A) (PC) are attractive since it is expected that they exhibit properties as good as those of PET/PC and PBT/PC blends, where compatibility exists thanks to their partial miscibility<sup>2–9</sup> and to the interchange reactions<sup>6,10–15</sup> that may occur in the melt state. Consequently, PTT/PC blends have also been the subject of attention. The phase structure,<sup>16–19</sup> transesterification reactions<sup>20–22</sup> and crystalline behavior<sup>17,19,23–25</sup> of PTT/PC blends have been studied. The blends were shown to be partially miscible<sup>16–19</sup> with different miscibility levels,<sup>16–22</sup> probably because of the different interchange reaction level attained.<sup>19–22</sup> The progressive development of interchange reactions gives rise<sup>20–22</sup> to the homogenization of the blends and eventually leads to single-phase materials. PC hinders PTT crystallization in the blends,<sup>17</sup> decreasing drastically the level of crystallinity<sup>23–25</sup> by means of interchange reactions, that decrease the crystallizable segment length of PTT.<sup>24,25</sup> These reactions should affect the phase structure and mechanical properties. However, to our knowledge, no work has been published on the mechanical properties of PTT/PC blends.

In this study PTT/PC blends were obtained by mixing in the melt state either by direct injection molding (DI blends), or mixed previously by extru-

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sion followed by injection molding (PM blends). The phase structure was studied by differential scanning calorimetry (DSC), dynamic mechanical analysis (DMA), and Fourier transform infrared spectroscopy (FTIR); the morphology was studied by scanning electron microscopy (SEM); the solid state characteristics through density and birefringence measurements; and the mechanical properties by tensile tests. The morphology was related to the phase behavior of the blends, and the mechanical properties were discussed in terms of phase structure and morphology.

## EXPERIMENTAL

The polymers used in this work were PTT kindly supplied by Shell Chemical (Corterra CP509200) and PC, (PC2200R), which was obtained from Idemitsu Petrochemical. The MFI values of PTT and PC were 19.2 g/10 min and 2.0 g/10 min, respectively. Both were measured at 270°C and with a 0.325 kg load (ASTM D-1238). Both polymers were dried before processing to avoid moisture-induced degradation reactions, the PTT for 4 h at 135°C and the PC for 12 h at 120°C.

The blends were obtained by two methods: premixing by extrusion followed by injection molding (PM), and direct blending-injection molding (DI). Premixing of PM blends was carried out in a co-rotating twin screw extruder-mixer (Collin ZK-25). The diameter and length to diameter ratio of the screws were 25 and 30 mm, respectively. The barrel temperature was 270°C and the rotation speed 30 rpm. The rod extrudates obtained were cooled in a water bath, pelletized and dried at 120°C for 12 h before injection molding. Injection molding was carried out in a Battenfeld BA 230E reciprocating screw injection molding machine a melt temperature of 270°C and a mold temperature of 15°C. The screw had a diameter of 18 mm, a L/D ratio of 17.8, compression ratio of 4 and helix angle of 17.8°. No mixing devices were present in the plasticization unit. The injection speed and pressure were 8 cm<sup>3</sup>/s and 1750 bar, respectively. Tensile (ASTM D638, type IV) and impact (ASTM D256) specimens were obtained.

The thermal behavior of the blends and of the neat components was studied by DSC using a Perkin-Elmer DSC-7 calorimeter. The samples were heated from 25°C to 280°C at 20°C/min. The crystallization and melting temperatures and heats were calculated from the maxima and from the areas of the corresponding peaks, respectively. The glass transition temperatures of the blends were determined from the first heating scan. The phase structure was studied by DMA using a TA Instruments Q800 DMA, which provided the loss tangent ( $\tan \delta$ ) values against temperature from -140 to 180°C at a heating rate of 4°C/min and a frequency of 1 Hz.

Fourier Transform Infrared (FTIR) spectra were obtained using a Nicolet Magna 560 spectrophotometer, both in solid specimens and in dissolved fractions obtained by solvent extraction. The theoretical FTIR spectra were calculated from the weighted addition of the FTIR spectra of the neat components for each composition. The specific volumes of the blends and the neat components were measured in a Mirage SD-120-L electronic densitometer with a maximum deviation of 0.0008 cm<sup>3</sup> g<sup>-1</sup>, using butyl alcohol as the immersion liquid. The birefringence was measured at room temperature in an Olympus BX40 microscope equipped with a compensator. Each birefringence value was obtained from an average of at least three measurements. The morphology of the cryogenically fractured tensile specimens was observed by SEM after gold coating. A Hitachi S-2700 microscope was used at an accelerating voltage of 20 kV. The melt flow indices (MFI) of the blends were measured (ASTM D1238) using a Ceast extrusion plastometer at 270°C and with a 0.325 kg load.

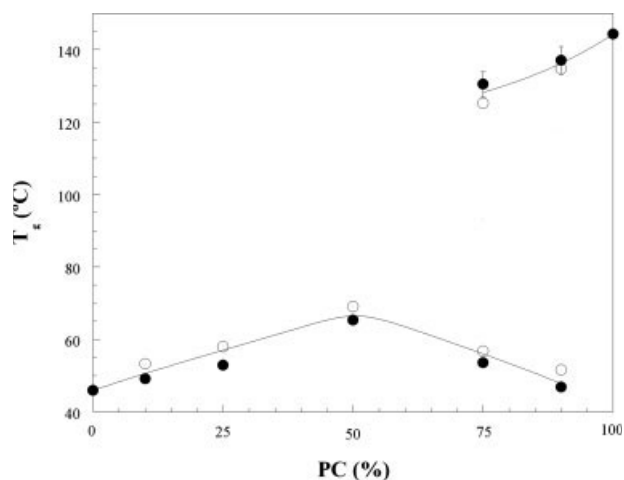
Tensile testing was carried out using an Instron 4301 machine at a cross-head speed of 10 mm/min, at 23 ± 2°C and 50 ± 5% relative humidity. The mechanical properties (tensile strength ( $\sigma_t$ ) and elongation at break ( $\epsilon_b$ )) were determined from the load-displacement curves. The Young's modulus was determined initially by means of an extensometer at a cross-head speed of 1 mm/min. The impact tests were carried out using a Ceast 6548/000 pendulum on notched specimens. The notches (depth: 2.54 mm and radius: 0.25 mm) were machined after molding. At least eight specimens were tested for each reported value in both the tensile and impact tests.

## RESULTS AND DISCUSSION

### Phase behavior

#### Amorphous phase

Figure 1 shows the glass transition temperatures of the blends ( $T_g$ ) measured by DSC against composition both, for the blends obtained by direct injection molding (DI blends) and, for the blends obtained with previous mixing (PM blends). The same plot obtained by DMA had similar characteristics; the values were, however, slightly higher, as expected from the DMA frequency used. As can be seen, the PC-rich blends show two  $T_g$ 's. The low temperature  $T_g$  appears at temperatures above that of the pure PTT, indicating the presence in the blends of a PTT-rich phase where some PC is present. The high temperature  $T_g$  decreases when the PTT content of the blend increases, indicating the presence of a PC-rich phase where some PTT is present. The PTT-rich blends, as well as the intermediate compositions,



**Figure 1** Glass transition temperature ( $T_g$ ) of PTT-rich phase and PC-rich phase of the PM (○) and DI (●) blends determined by DSC.

showed a single  $T_g$  located at temperatures between those of the pure components, which increases with the PC content in the blend. This indicates the presence of a single amorphous phase and suggests miscibility, at least at the detection level characteristic of DSC (and DMA). A similar change of the  $T_g$  with composition was observed in PET/PC blends<sup>26,27</sup> where full miscibility at PET contents higher than 60% occurred and was attributed to strong interactions.

Different phase behaviors of the PTT/PC blends have been reported previously in the bibliography. Partial miscibility,<sup>16–18</sup> as well as, full immiscibility<sup>20</sup> throughout the whole composition range were reported. However, upon annealing at high temperatures the presence of a single phase was reported.<sup>16,20–22</sup> This clearly indicates the presence of interchange reactions in the melt state. These reactions could also take place in the blends of this study and will be studied later. Tentatively, the differences in phase behavior between the blends of this study and those of the bibliography are attributed to the different extent of the interchange reactions consequence of the different processing methods and conditions used.

The compositions of the two phases of the biphasic blends were estimated from the corresponding  $T_g$  values of each phase using the Fox equation (1).<sup>28</sup>

$$\frac{1}{T_g} = \frac{\omega_{\text{PTT}}}{T_{g,\text{PTT}}} + \frac{\omega_{\text{PC}}}{T_{g,\text{PC}}} \quad (1)$$

where  $T_{g,\text{PTT}}$  and  $T_{g,\text{PC}}$  are the glass transition temperatures of neat PTT and PC, respectively, and  $\omega_{\text{PTT}}$  and  $\omega_{\text{PC}} = 1 - \omega_{\text{PTT}}$  the weight fractions of both components. The calculated  $\omega_{\text{PTT}}$  and  $\omega_{\text{PC}}$  values indicated that in the 25/75 blends for instance, the

PTT-rich phase contained 21 and 28% of PC, respectively, in DI and PM blends, and the PC-rich phase contained respectively 5 and 7% PTT.

As can also be seen in Figure 1, the  $T_g$ 's of the PM blends are more shifted from the  $T_g$ 's of the corresponding neat components than those of the DI blends. The difference is small, but significant, because it took place at all compositions and appeared also in the  $T_g$ 's measured by DMA. The differences are easily assessed in Table I. The  $T_g$  values of the PTT-rich compositions of the DI method are slightly lower. This indicates a lower PC content than in the correspondent PM blends. This lower PC content can be equilibrated only through the presence of undetectable amounts of unmixed PC-rich phase consequence of a no fully perfect mixing. In PM PC-rich blends, the smaller distance between the  $T_g$  of the PTT-rich phases and those of the PC-rich phases indicates that both phases have a more similar composition, in agreement with the higher contact time in the melt state of the PM method.

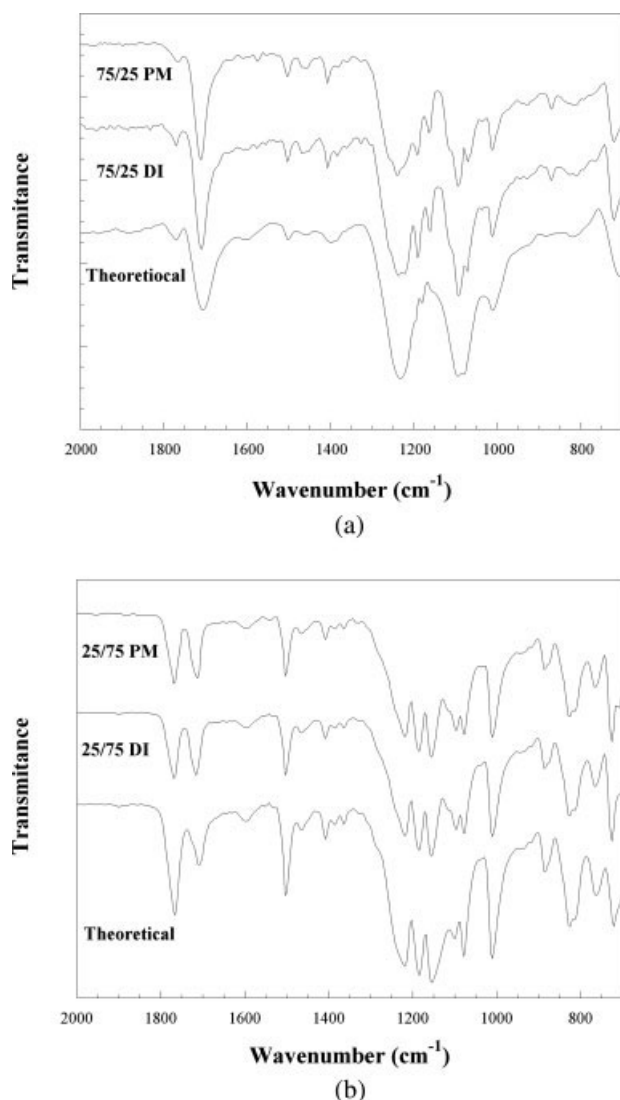
#### Interchange reactions

The production of copolymers during mixing in the melt state that consequently modify the phase behavior of the blend has been observed, among others, in PTT/PC blends<sup>16,20,21</sup> as well as in blends of PC with polymers similar to PTT, such as PET<sup>29–31</sup> and PBT.<sup>10,32,33</sup> To test the possible presence of copolymers in the blends of this study, the 75/25 and 25/75 blends were analyzed by FTIR. The results were compared with the theoretical results obtained from the combination of the spectra of the pure components and are shown in Figure 2.

As can be seen in the experimental spectra of the 75/25 blends [Fig. 2(a)] obtained by the two methods, a new band that was not present in the theoretical spectrum appeared at 1070  $\text{cm}^{-1}$ . This band also appeared in PBT/PC blends<sup>34</sup>; it was attributed to a complex vibration of the *para*-disubstituted phenyl to the right of the ester structure influenced by the neighboring  $-\text{COO}-$  group, and proves the production of copolymers.

**TABLE I**  
 **$T_g$ 's of the DI and PM Blends Obtained by Means of DMA**

PTT/PC	$T_{g(\text{PTT})}$ DI	$T_{g(\text{PTT})}$ PM	$T_{g(\text{PC})}$ DI	$T_{g(\text{PC})}$ PM
0/100	–	–	144.5	144.5
10/90	47.0	51.5	137.0	135.0
25/75	53.5	57.0	130.5	125.0
50/50	65.5	69.0	–	–
75/25	53.0	58.0	–	–
90/10	49.0	53.0	–	–
100/0	46.0	46.0	–	–



**Figure 2** Experimental and theoretical FTIR spectra of PTT/PC blends at PC contents of (a) 25% and (b) 75%.

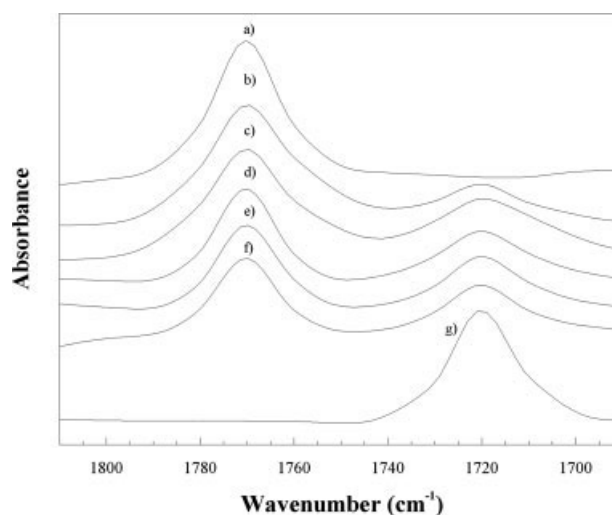
In the case of the 25/75 blends of Figure 2(b), no difference was observed between the experimental and theoretical spectra. This, however, does not rule out the presence of reactions, which could have been produced to a small extent, rendering them undetectable in the whole blend. For this reason, the blends were treated with chloroform, which is a selective solvent for PC. Consequently, provided reactions do not occur, the FTIR spectrum of the soluble fractions should correspond to PC only. The spectra of the soluble fractions of DI blends (those of the PM blends were similar) are shown in Figure 3. As can be seen, there is a band at  $1720\text{ cm}^{-1}$  that corresponds to the carbonyl stretching vibration band of the PTT. This proves the presence of PTT in the soluble fraction, the presence of copolymers and, to a minor extent, the occurrence of reactions also in the 25/75 composition.

To find out whether our blends are reacted or mixed, we will compare the results of this study with those of the bibliography. In previous works on PTT/PC blends<sup>16,17,20</sup> the  $T_g$  of the PTT-rich blends was  $7\text{--}14^\circ\text{C}$  above that of pure PTT in the absence of reactions. If we compare this change with that of this study (up to  $22^\circ\text{C}$  depending on composition), we can see that in this case the change is larger. Provided mixing in the cited references was effective, the large  $T_g$  change of this study testifies to the occurrence of chemical reactions. On the basis of the respective changes of  $T_g$ , we can conclude that we have reacted copolymers present. Therefore, the migrated components are both reacted and mixed.

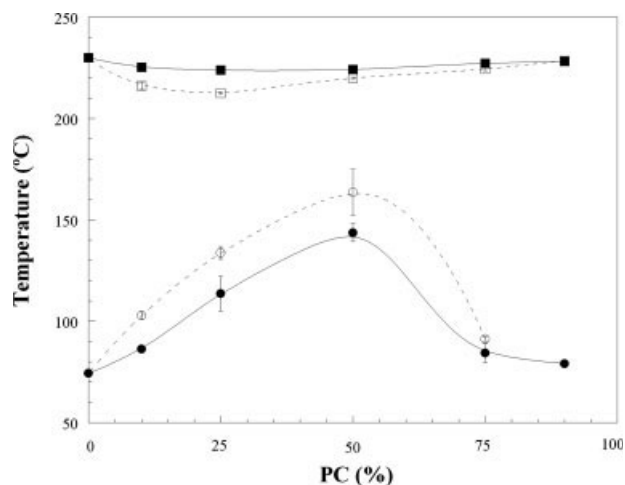
### Crystalline phase

The melting ( $T_m$ ) and the crystallization ( $T_c$ ) temperatures of both the DI and PM blends and of the pure PTT are shown in Figure 4 as a function of composition. It can be seen that the  $T_m$  of the PTT-rich phase decreased in presence of PC, mainly at high PTT contents. This indicates a decrease in the length of the crystalline PTT segments that is consistent with the occurrence of interchange reactions and the presence of miscibility<sup>32,35</sup> mentioned in the previous section. This shorter segment length leads to a more imperfect crystalline phase and therefore, to a smaller  $T_m$ . Further, we observe a greater decrease in  $T_m$  in the PM blends. This agrees with previous observations in PET/PC blends,<sup>32,33</sup> and with the larger extent of the interchange reactions present in PM blends.

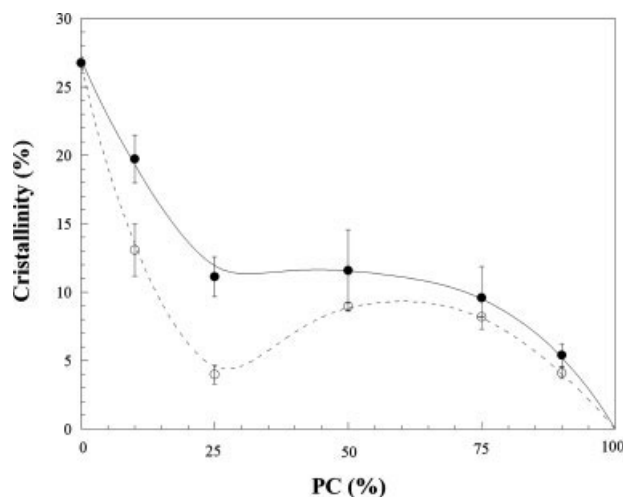
In the PTT-rich compositions of Figure 4, the  $T_c$  increased with the PC content, and then decreased.



**Figure 3** FTIR spectra of chloroform-soluble fractions of the PTT/PC DI blends, (b) 10/90, (c) 25/75, (d) 50/50, (e) 75/25 and (f) 90/10; and of the neat components (a) PC and (g) PTT.



**Figure 4** Crystallization (DI: ● y PM: ○) and melting (DI: ■ y PM: □) temperatures of the PTT rich phase as a function of the PC content.



**Figure 5** Crystallinity percentage of PTT/PC blends (DI: ● y PM: ○) versus PC content.

This unusual behavior observed before<sup>17,26</sup> is similar to the behavior of the low temperature  $T_g$ , and is attributed to the limited miscibility of the system. The  $T_c$  of the PM blends appeared at higher temperatures, indicating a hindrance in the crystallization of PTT, which also agrees with the larger extent of the interchange reactions present in PM blends.

The crystalline content of the blends with respect to composition is shown in Figure 5. As can be seen, in the intermediate and PTT-rich blends the crystalline content of the blends lies below linearity. The crystalline content of PTT in the blends was smaller than that of the pure PTT. Above 50% PC, however, the crystalline content was slightly higher than that expected by simple linear addition of the crystallinity of the two components, indicating that the crystalline content of PTT in the blends was similar or slightly higher than that of the pure PTT. The lower crystalline content of PTT in the PTT-rich blends is consistent with the hindering of crystallization due to the presence of miscibilized PC. The fact that the crystallinity of PTT is higher in the PC-rich blends than in pure PTT is difficult to explain, but a similar behavior has been observed previously in PBT/poly (arylate of bisphenol A) PAI blends.<sup>36</sup>

### Morphology

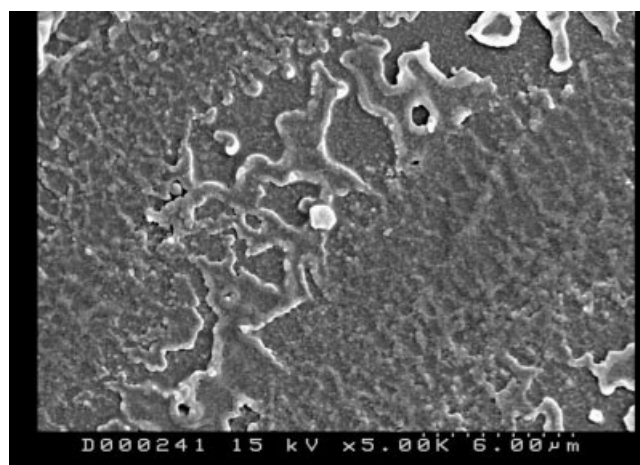
The cryofractured surface of the PC-rich blends was observed by SEM. Surprisingly, no dispersed phase was observed. As the presence of two phases has been demonstrated, this indicates that fracture was cohesive and, therefore, the adhesion level was high. Additional information concerning the morphology of the dispersed phase of the PC-rich blends could be obtained by selective chemical attack of PTT; however, no selective attack agent was found.

Selective chemical attack by means of diethylene triamine was possible, however, for PC. This provides additional experimental support to the presence of miscibility in PTT-rich blends. This is because it is not completely clear which technique, DMA or SEM, is the most sensitive to the presence of very small sizes of the dispersed phase. The attacked surface of the 75/25 blend is shown in Figure 6.

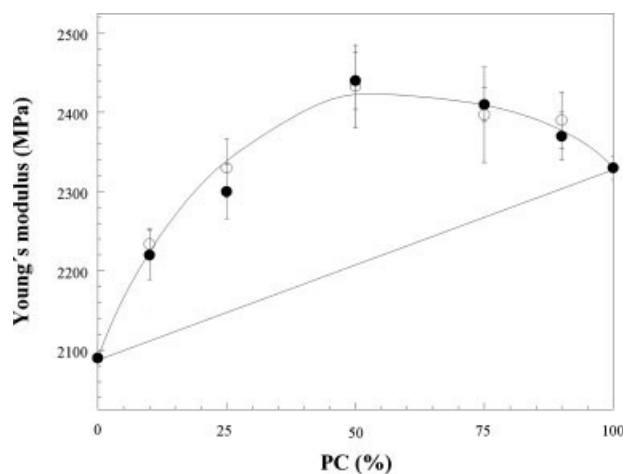
As can be seen, no dispersed phase was observed at the maximum magnification, compatible with the irregular surface. This probes that, if a dispersed phase not observable by either DSC or DMA exists, it must be smaller than 0.1  $\mu\text{m}$ .

### Mechanical properties

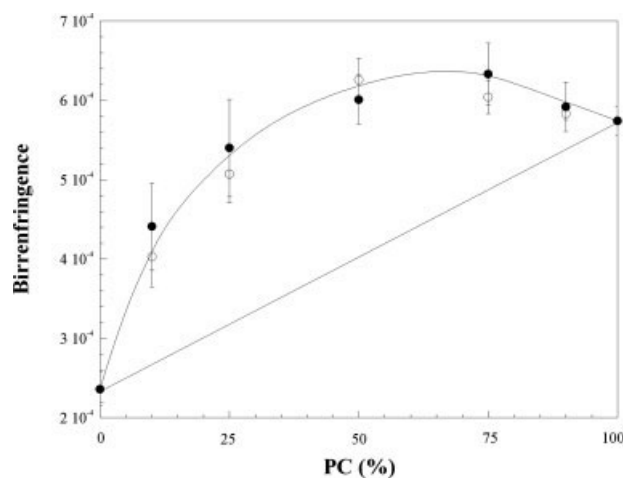
The Young's modulus of the blends is shown in Figure 7 versus composition. It can be seen that the



**Figure 6** SEM micrograph of the etched surface of a cryofractured tensile specimen of 75/25 DI PTT/PC blend.



**Figure 7** Young's modulus of PTT/PC blends (DI: ● and PM: ○) versus PC content.



**Figure 8** Birrefringence of PTT/PC blends (DI: ● and PM: ○) versus PC content.

modulus of elasticity of the blends exhibits an important synergism throughout the whole composition range. Moreover, the modulus values did not depend on the processing method. This was expected, since the modulus should be independent of interfacial adhesion.<sup>31</sup> Synergisms in the modulus of elasticity, are often seen in miscible blends, such as PBT/PAr<sup>36</sup> or PET/PBT.<sup>37</sup> In the PTT//PC blends of the present work, the synergism also occurs in the partially miscible PC-rich region. A synergistic modulus behavior has also been found in other partially miscible blends.<sup>3,38</sup>

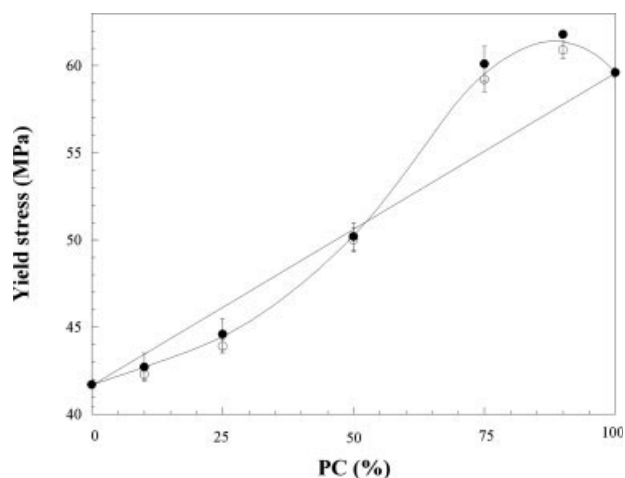
The behavior of the modulus of elasticity can be mainly due to (a) a densification of the amorphous phase of the blends due to specific interactions between their components, i.e., a negative volume of mixing, (b) a higher degree of crystallinity of PTT in the blends, or (c) a higher orientation in the blends than in the pure components. The changes observed in crystallinity upon blending are too small to explain the modulus behavior (Fig. 5); therefore, this parameter can be ruled out as the reason for the observed behavior. The change of the specific volume values was close to the experimental error, so it cannot explain the large change in modulus observed.

The orientation of the blends (as measured by birefringence) as a function composition is shown in Figure 8. It can be observed that the orientation is much higher in the blends than predicted by simple linear addition. Moreover, the increase in orientation is the highest where the synergism of modulus was also the largest, without any significant difference observed between the DI and PM blends. It is thus clear that the higher orientation in the blends is the main reason for the observed synergism in the modulus. This higher orientation in the blends is a consequence of the difference in viscosity of the two com-

ponents of the blends; this will be seen experimentally after because of their clearly different MFI.

The yield stress of the blends is shown in Figure 9 against composition. We can see a different behavior from that of the modulus. Usually, the behavior of both properties is the same,<sup>39</sup> although some exceptions have been reported,<sup>40–42</sup> because both are basically affected by the same parameters. In this case, the substantial decrease in the crystallinity of PTT upon blending in the PTT-rich blends (it was half that of the neat PTT in the 75/25 blend), should lead to a decrease in the yield stress. Overall, the plots of the yield stress and the crystallinity are rather similar. This allows us to attribute the variations of the yield stress of the blends mainly to the change in crystallinity of the PTT.

The elongation at break of the DI and PM blends is shown in Figure 10 as a function of composition. As can be seen, all the blends are ductile, as

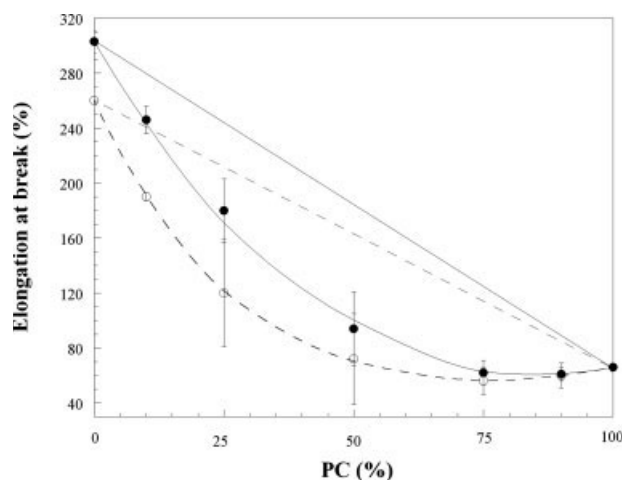


**Figure 9** Yield stress of PTT/PC blends (DI: ● and PM: ○) versus PC content.

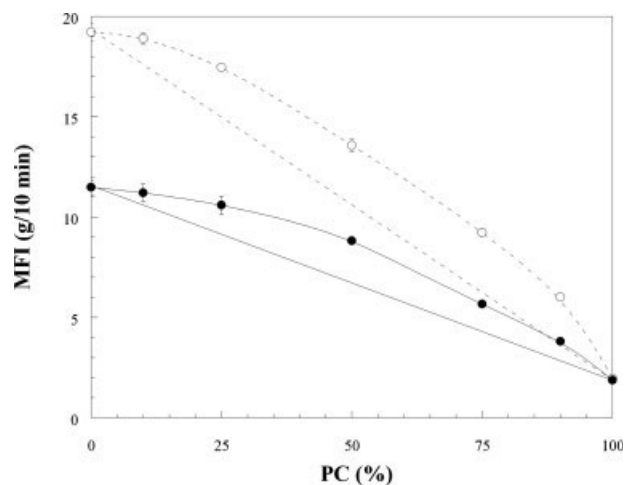
expected from the good interfacial adhesion observed, but there is a general decrease in elongation at break after blending. This cannot be attributed to either orientation or crystallinity, because the observed changes were too small to clearly modify the elongation at break. The decrease in elongation at break in miscible blends or in biphasic blends where both components are present in the two phases, thus assuring compatibility, is usually due to a decrease in the free volume<sup>43–45</sup> induced by blending. However, the change in the specific volume was close to the experimental error. The decrease in elongation at break, with respect to that given by simple linear addition, occurred in all compositions indicating that there must be another fact influencing elongation at break.

When the reprocessing ability of PTT was studied, a decrease in elongation at break was observed.<sup>46</sup> Similarly, the elongation at break of PET/PC blends also decreased when extruded.<sup>31</sup> Since in both cases the decrease was attributed to a decrease in the molecular weight of the polyester, this possibility was tested in the blends of this study by measuring the MFI of the blends.

The composition dependence of the MFI in the DI and PM blends is plotted in Figure 11. As can be seen, the MFI of the blends is higher than that expected by simple linear addition of the MFI of the pure components. This experimental viscosity decrease indicates degradation.<sup>31,46</sup> Degradation is also deduced from the higher decreases in elongation at break observed in the harsher PM processing. To find out what is the main component suffering degradation, we realize that the harsher PM processing procedure influences neither the MFI nor the elongation at break of PC. However, it clearly affects the MFI and the elongation at break of PTT. Therefore, degradation of PTT occurs during processing



**Figure 10** Elongation at break of PTT/PC blends (DI: ● and PM: ○) versus PC content.



**Figure 11** MFI of PTT/PC blends (DI: ● and PM: ○) versus PC content.

and, therefore, it is proposed as responsible of the observed behavior of the elongation at break.

## CONCLUSIONS

The PTT-rich PTT/PC blends of this study are monophasic and probably miscible. The PC-rich blends are partially miscible, with a higher content (roughly 25% in the 25/75 blend) of the other component in the PTT-rich phase than in the PC-rich phase (5% in the same blend). Since interchange reactions occurred during processing, the blends are not only miscible or partially miscible, but also partially reacted. The extent of the reaction is greater in PM blends. Dispersed particles were not visible with SEM in the PC-rich blends, indicating that fracture was cohesive and that the interphase adhesion was significant.

The modulus of elasticity of the blends showed a clear synergism that was mainly attributed to the higher orientation in the blends as compared to the neat state. This synergism opposes the effects related to changes in the specific volume and crystallinity, which were not large enough to influence the modulus significantly. The change in crystalline content of PTT upon blending did influence, however, the yield stress values.

All the blends were clearly ductile, although a decrease in the elongation at break was observed upon blending, mainly in PM blends. This is probably the result of some degradation of the PTT. The high elongation at break was a consequence of the cohesive fracture observed by SEM and of the monophasic or partially miscibilized structure that assured good interface conditions when dispersed phases were present.

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