Structure and Mechanical Properties of Poly(trimethylene terephthalate)/Poly(hydroxy ether of bisphenol A) Blends

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ABSTRACT: Compatible poly(trimethylene terephthalate) (PTT)/poly(hydroxy ether of bisphenol A) (Phenoxy) blends were obtained by direct injection molding throughout the composition range. Two amorphous phases with minor amounts of the other component were found in the blends. Reactions occurred in PTT-rich blends. By comparing the miscibility level of these blends with that of other blends based on polyalkylene terephthalates, it is proposed that a miscibility limit delimited by a 3/1 methylene–carbonyl ratio in the polyalkylene terephthalate exits in these blends. The synergism in the Young's modulus of the blends is discussed as a consequence of the changes in the crystallinity of PTT, the specific volume and the orientation produced by blending. Ductility is approximately proportional to blend composition, indicating compatibility, and is attributed to the combined effects of a small particle size and a good adhesion level, the latter being a consequence of the partially miscible nature of the blends. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 102: 3246–3254, 2006

Key words: polyethers; polyesters; blends; mechanical properties; melt

INTRODUCTION

The development of new polymeric materials currently follows two main paths: (i) the synthesis of new polymeric structures and (ii) the modification of existing polymers either by incorporating fillers or reinforcements or by blending with other polymers. These materials exhibit enhanced properties that open up new applications and allow the substitution of traditional materials by polymeric materials. Moreover, the two development paths may be combined. This is because when a new polymer is commercialized, its blends with the second components, which are expected to offer benefits, are usually studied and characterized.

Poly(trimethylene terephthalate) (PTT), a relatively novel thermoplastic polymer, belongs to the aliphatic–aromatic polyalkylene terephthalate family, which also includes the well-known and widely applied poly (ethylene terephthalate) (PET) and poly (butylene terephthalate) (PBT). PTT shows, in addition to the chemical resistance of thermoplastic semicrystalline polyalkylene terephthalates, resilience and elastic recovery similar to those of nylon 6,6.[1] Moreover, it has very good dyeability characteristics, which is a desirable property in textile applications. Blending PTT with other thermoplastics could combine these properties with those of the second blend component. However, because of its relatively recent commercialization only a few thermoplastics, mainly polyalkylene terephthalates and related polymers, have been blended with PTT. Thus, PTT was found to be (a) fully miscible with PET,[2] PBT,[3] poly(ethylene naphthalate),[4] PETG,[5] and poly(ether imide) (PEI),[6] (b) partially miscible with a polyarylate of bisphenol A and isophthalic/terephthalic acids (PAr)[7] and bisphenol A polycarbonate (PC),[8] and (c) immiscible with polystyrene (PS)[9] and ethylene–propylene–diene rubber (EPDM).[10] In the case of PTT/PC blends, the partial miscibility seems to be mainly due to the presence of interchange reactions. Ternary PET/PTT/PBT[11] and quaternary PET/PTT/PBT/PEI[12] blends have also been obtained and characterized.

The poly(hydroxy ether of bisphenol A) (phenoxy) is an amorphous thermoplastic characterized by relatively high ductility and toughness, as well as good oxygen barrier behavior. Its chemical structure includes a lateral hydroxyl group in each repeating unit, which is able to establish specific interactions with proton-acceptor polymers and is also able to react with condensation polymers such as polyalkylene terephthalates, polyamides, or PC. Miscible phenoxy blends include those with PBT,[13–18] poly(ε-caprolactone),[19] poly(methyl methacrylate),[20] Hytrel,[21] and poly(ethylene oxide).[22] Partially miscible blends

include those with poly(ethyl methacrylate).[23] Immiscible blends of phenoxy with PET,[14,18,24] polyamide 6,[25] PC,[26] poly(methylene oxide),[27] polypropylene,[28] polyethylene,[29] SAN,[30] PAr,[31] and ABS[32] have been reported. Ternary blends were studied, in which phenoxy was a component[33] or compatibilizer for polyamide 6/PBT[34] and polyamide 6/PET[35] blends. Phenoxy has also been incorporated into the matrix of supertough PBT-based blends with a poly(ethylene-octene) copolymer.[36]

The reasons for the miscibility state of polyalkylene terephthalates with other polymers, such as PEI, PAr, and phenoxy, are not clear. Thus, phenoxy was seen to be miscible with poly(hexamethylene terephthalate) and PBT,[13–18] while it was seen to be immiscible with a laboratory synthesized PTT [poly(1,3-propylene terephthalate)], poly(1,2-propylene terephthalate), and PET.[14,18,24] It was expected that the pendant hydroxyl group of the phenoxy interacted with carbonyl of the polyalkylene terephthalate to promote miscibility. However, the miscibility was not observed in all the systems, indicating that the specific interactions between hydroxyl groups in phenoxy and carbonyl groups in the polyalkylene terephthalate are not the only factor determining miscibility between phenoxy and polyalkylene terephthalates.

In a study on polyalkylene terephthalates/PC blends,[37] it was stated that the aliphatic carbon/ester group ratio of the polyalkylene terephthalate affected the miscibility of the system. PET, PTT, and PBT have only slight structural differences, which can be used to gain an insight into the role that this ratio has on the miscibility between polyalkylene terephthalates with polymers such as PEI, PAr, etc. Thus, taking into account the miscibility of PBT/phenoxy blends and the immiscibility of the PET/phenoxy system, it is interesting to study in depth the miscibility state of PTT/phenoxy blends, to determine whether the ratio in the polyalkylene terephthalates is also the reason that determines their miscibility. Additionally, the characterization of the structure and properties of PTT/phenoxy blends should also be useful from the point of view of the development of new polyalkylene terephthalate-based materials. This is because the T_g behavior of the laboratory synthesized PTT/phenoxy blends was reported,[14] but to our knowledge, neither the possibility of reactions nor the morphology nor the mechanical properties have been studied.

In the present work, the solid-state structure and the mechanical properties of PTT/phenoxy blends have been studied across the whole composition range. The blends were prepared in the melt state by a direct mixing-injection molding procedure. The miscibility level and the phase behavior were studied by differential scanning calorimetry (DSC) and dynamic-mechanical analysis (DMA), and the morphology was observed by scanning electron microscopy (SEM). Tensile and notched impact tests were performed to determine the mechanical properties of the blends.

EXPERIMENTAL

The polymers used in this work were commercial products. PTT was kindly supplied by Shell Chemical under the trade name Corterra CP509200. It has an intrinsic viscosity of 0.92 dL/g, as measured in a phenol/1,1,2,2,-tetrachloroethane (50:50) mixture at 30°C, and an MFI of 10.5 g/10 min at 240°C and under a load of 0.320 kg (ASTM D1238). Phenoxy (Paphen PKHH) was obtained from Phenoxy Specialties (Union Carbide), and it had a viscosity of 525–715 cP in a 20% cyclohexanone solution at 25°C and an MFI of 2.4 g/10 min (240°C and 0.320 kg load). Both polymers were dried before processing to avoid moisture-induced degradation reactions, the PTT for 4 h at 135°C and phenoxy for 12 h at 80°C.

The PTT/phenoxy blends were directly melt-mixed and injection-molded using a Battenfeld BA230E reciprocating screw injection molding machine at a melt temperature of 230°C and a mold temperature of 15°C. The screw had a diameter of 17.8 mm and an L/D ratio of 17.8. No mixing devices were present in the plasticization unit. The injection speed and pressure were 5 cm³/s and 2500 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 differential scanning calorimetry (DSC) using a Perkin–Elmer DSC-7 calorimeter. The samples were heated from 15 to 260°C at 20°C/min, cooled at the maximum speed provided by the calorimeter (∼ 100°C/min), and were subjected to a second heating scan under the same conditions as the first. The crystallization and melting temperatures and heats were calculated from the first heating scan, at the maxima and from the areas of the corresponding peaks, respectively. The glass transition temperatures of the blends were determined from the second heating scan, to allow a better observation. The phase structure was indirectly studied by dynamic mechanical analysis (DMA) using a TA Instruments Q800 DMA, which provided the loss tangent (tan δ) values against temperature. The temperature scans were carried out from −110 to 130°C at a heating rate of 4°C/min and a frequency of 1 Hz.

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 cm³ g⁻¹, using butyl alcohol as the immersion liquid. Fourier transform infrared (FTIR) spectra were obtained using a Nicolet Magna 560 spectrome-
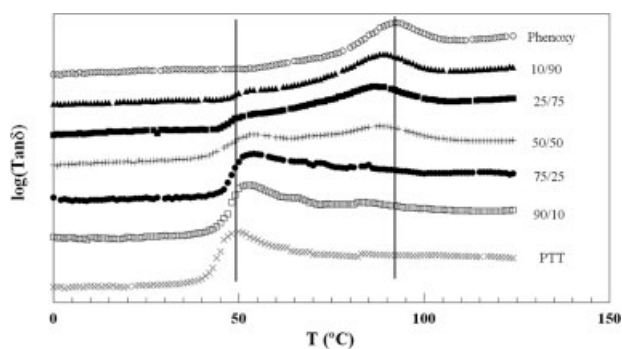
Figure 1 DMA log (tan δ) versus temperature plots of PTT/phenoxy blends. The curves are shifted on the vertical axis to aid clarity.
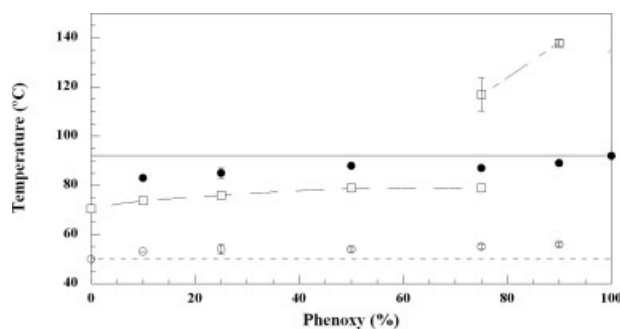

Figure 2 Thermal transitions of PTT/phenoxy blends as a function of the phenoxy content. Glass transition temperature (T_g) of PTT-rich phase ○ and phenoxy-rich phase ●, obtained by DMTA, and the crystallization temperature (T_c) □, obtained by DSC.

ter, both in solid specimens and in dissolved fractions obtained by solvent extraction with chloroform in which phenoxy is soluble and PTT is not. The theoretical FTIR spectra were calculated from the weighted addition of the FTIR spectra of the neat components for each composition.

Tensile tests were carried out using an Instron 4301 instrument at a cross-head speed of 10 mm/min and at $(23 \pm 2)°C$. The mechanical properties (Young's modulus, E; yield stress, σ_y; and stress and strain at break, σ_b and ε_b respectively) were determined from the load-elongation curves. 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.

The morphology of the cryogenically fractured tensile specimens was studied by scanning electron microscopy (SEM) after gold coating. A Hitachi S-2700 microscope was used at an accelerating voltage of 15 kV. The phenoxy-dispersed phase was selectively dissolved from some samples with chloroform for 4 h to allow a better observation.

RESULTS AND DISCUSSION

Phase behavior

The phase behavior of the blends was studied both by DSC and DMA. The tan δ–temperature scans of the blends and of the neat components are shown in Figure 1, and the plots of the glass transition temperature (T_g) and crystallization temperature (T_c) of the blends against composition are shown in Figure 2. The T_g's were determined at the maxima of the peaks by DMA and the T_c's by DSC. The DSC results from the second heating scan are shown in Figure 3.

As can be seen in Figure 1, two tan δ peaks appeared in the blends. The minor peak was difficult to observe in compositions containing very low levels of the minor component. Both peaks were close to those of the corresponding neat components and showed a slight tendency to approach to each other as seen both in Figures 1 and 2. The T_g's measured by DSC showed the same trends as those obtained by DMA. These results indicated the presence of both a phenoxy-rich and a PTT-rich phase in the blends; in both phases some amount of the other component was present. Two unchanged T_g's have been observed previously[14] in laboratory-synthesized PTT/phenoxy blends, reported in a short note on the phase behavior of polyalkylene terephthalates/phenoxy blends as observed by DSC. The compositions of both phases of the blends of this study were estimated by means of the Fox equation [eq. (1)].

$$\frac{1}{T_g} = \frac{\omega_{PTT}}{T_{g,PTT}} + \frac{\omega_{phenoxy}}{T_{g,phenoxy}} \tag{1}$$
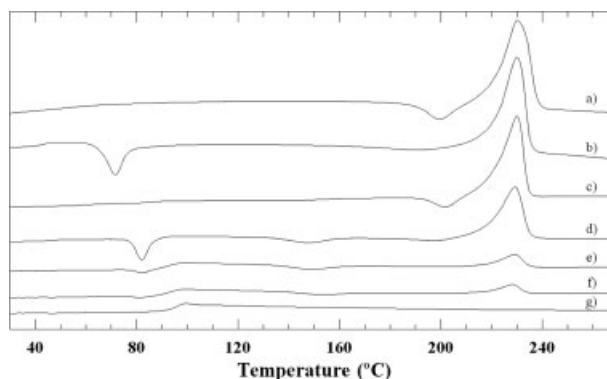

Figure 3 DSC second heating scan thermograms of (a) neat PTT and (g) phenoxy, and blends at phenoxy contents of (b) 10%, (c) 25%, (d) 50%, (e) 75% and (f) 90%.

TABLE I
PTT Weight Fraction in the PTT-Rich and Phenoxy-Rich Phases of PTT/Phenoxy Blends

PTT/phenoxy composition	PTT weight fraction	
	PTT-rich phase	Phenoxy-rich phase
100/0	1.00	–
90/10	0.90	0.19
75/25	0.90	0.14
50/50	0.89	0.12
25/75	0.86	0.10
10/90	0.83	0.06
0/100	–	–

From the T_g's determined by DMA, where T_g, $T_{g,PTT}$, and $T_{g,phenoxy}$ are the T_g values of the blend and of the two pure components, and ω_{PTT} and $\omega_{phenoxy}$ are the weight fractions of both components. The calculated results are reported in Table I. As can be seen, the presence of the minor component in each phase was relevant.

The mixed nature of the two amorphous phases of the blends could be due to partial miscibility, interchange reactions during processing, or both. Reactions producing copolymers of both components are likely to take place as they have been observed in other polyalkylene terephthalate/phenoxy systems in PET/phenoxy[24] and PBT/phenoxy[16,17] blends. To test for reactions, the 75/25 and 25/75 blends were analyzed by FTIR. Their spectra are shown, respectively, in Figures 4(a) and 4(b). As can be seen, in the phenoxy rich blend, in Figure 4(b) a new stretching band of C=C bond at 1650 cm^{-1} appeared and the stretching band of the ether group of phenoxy at 1300 cm^{-1} almost disappeared. As these changes in the chemical structure appear to be related to phenoxy and as they do not appear in phenoxy poor blends, they are tentatively attributed to degradation of phenoxy. As can also be seen, the experimental and theoretical spectra of the PTT-rich blends were practically identical. As this result does not clarify whether the blends are reacted or not, they were placed in chloroform, and the soluble fraction was studied by FTIR (Fig. 5). As phenoxy is soluble and PTT is not, the soluble fraction should have only phenoxy if reactions did not occur. The spectra of the chloroform-soluble fractions of the 90/10 and 75/25 blends showed a clear stretching band of PTT at 1720 cm^{-1} of higher intensity in the case of 75/25 blend. In the 50/50, 25/75, and 10/90 blends, no band that could correspond to PTT was seen. This indicates that reactions occurred only in the case of PTT-rich blends. The reacted products should only be present in the PTT-rich phase of the PTT-rich blends, because they were not detected in the phenoxy-rich compositions. Therefore the PTT/phenoxy blends are partially miscible, comprising two amor-
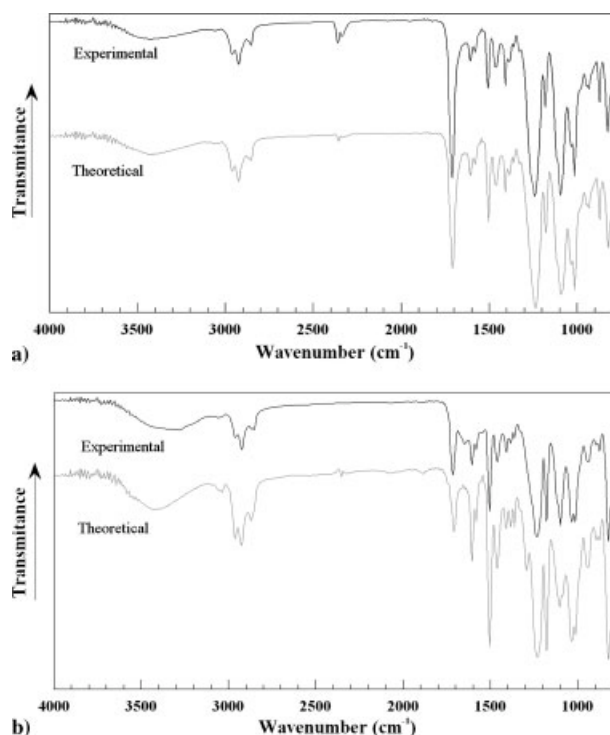

Figure 4 Experimental and calculated FTIR spectra of PTT/phenoxy blends at phenoxy contents of (a) 25% and (b) 75%.

phous phases rich in PTT and phenoxy, in which small amounts of phenoxy and PTT, respectively, were present. In PTT-rich blends, the presence of the minor component was, at least partially, as a reacted copolymer.

The miscibility level of these PTT/phenoxy blends, together with that of the PBT/phenoxy and PET/phenoxy blends observed in previous studies, helps in understanding the reasons for miscibility in polyalkylene terephthalates/phenoxy blends. Thus, PBT is fully miscible[13–18] with phenoxy, the PTT of this study is partially miscible, and PET is immiscible
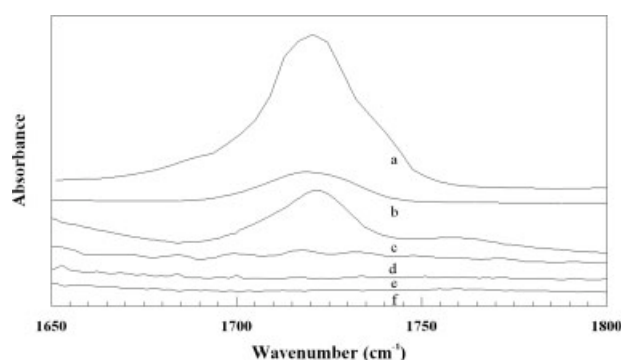

Figure 5 FTIR spectra of (a) neat PTT and chloroform-soluble fractions of the blends (b) 90/10, (c) 75/25, (d) 50/50, (e) 25/75, and (f) 10/90.

with phenoxy.[14,18,24] Thus, it appears that the miscibility level decreases with the increase in carbonyl group density in the polyalkylene terephthalates. Similar behaviors were observed both in polyalkylene terephthalates/PAr and polyalkylene terephthalates/PEI blends. In the blends with PAr, PBT[38] is miscible, while PTT[7] and PET[39] are partially miscible. In blends with PEI, PBT[40,41] and PTT[6] are fully miscible, while PET[42,43] is partially miscible. Thus, although there are slight differences depending on the nature of the second component, it appears that a miscibility limit appears to exist in blends based on polyalkylene terephthalates. This limit is close to the methylene/carbonyl group 3 : 1 ratio in the diol component of the polyalkylene terephthalate.

As can also be seen in Figure 2, the neat PTT and most of the blends showed a PTT cold-crystallization exotherm (represented as T_c) that increased slightly with the phenoxy content. This slight increase indicated a lack of strong interactions between PTT and phenoxy that agrees with the partial miscibility of the blends. This is because, as is usually found in immiscible blends, the T_c of PET/phenoxy blends[18] was independent of composition; and as is typical in miscible blends, that of PBT/phenoxy clearly increased with the phenoxy content. The 25/75 blend showed a second small exotherm at ∼ 120°C, indicating that PTT crystallized from both the amorphous phases of the blends. Finally, the 10/90 blend showed only a high temperature exotherm at ∼ 140°C, indicating a clear hindering of the crystallization process, which was expected, due to the large phenoxy content of the PTT-rich phase of this composition. The melting temperature of PTT remained practically constant with composition, as expected from the low miscibility level. This indicated that the crystalline perfection was basically the same in the blends as in the neat polymer. Although it was also seen in miscible PBT/phenoxy blends,[18] this is a usual behavior in immiscible and partially miscible systems.[25,43]

The crystallinity of the PTT in the blends was calculated from the experimental melting and crystallization heats in the first DSC scan and the melting heat of 100% crystalline PTT ($\Delta H_m = 145$ J/g).[44] It is plotted in Figure 6 against composition of the blend. The crystalline content was close to 24% in pure PTT and in the PTT-rich blends and ranged from 29% to 32% in the intermediate and phenoxy rich compositions. As can be seen in Figure 6, the crystallinity of the blends showed small positive deviations from the linear sum of the neat components. This indicates that although phenoxy hinders the crystallization kinetics of PTT, the final PTT crystallinity attained is slightly higher in the blends than in pure PTT. A qualitatively similar behavior has been found in other partially or fully miscible systems containing a crystallizable thermoplastic polyalkylene ter-
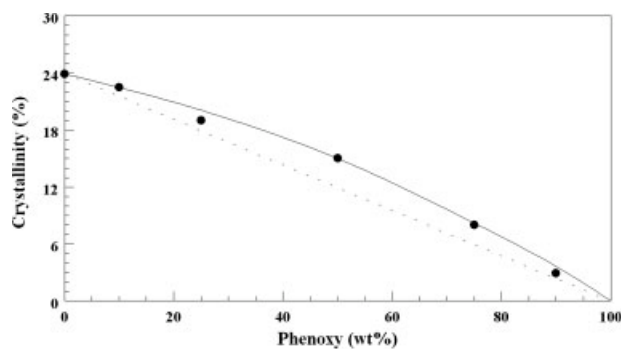

Figure 6 Crystallinity percentage of PTT in the blend versus phenoxy content.

ephthalate, such as PET/PAr,[39] PBT/PAr,[38] and PBT/phenoxy.[16]

Morphology

When the cryogenic fracture surfaces of the blends were observed by SEM, no dispersed phase was observed. As two phases were detected by DMA, this indicated that fracture was cohesive and, therefore, that the interfacial adhesion was significant. To reveal the morphology of the dispersed phase, the phenoxy phase in the fracture surfaces was dissolved with chloroform. Figure 7 shows the SEM photomicrographs of the PTT-rich and 50/50 blends. The phase inversion must be between 50 and 75% of phenoxy, because holes were seen in the 50/50 blend and the 25/75 blend was completely disintegrated. As can be seen in the 90/10 and 75/25 blends [Figs. 6(a) and 6(b) respectively), the dispersed holes and the corresponding dispersed phase were homogeneously distributed in the PTT matrix. Moreover, the particle size was small; typically 0.3 μm in the 75/25 composition and similarly in the 90/10 blend. The similar particle size of the 75/25 compared to that of the 90/10 blend was unexpected, because the particle size in polymer blends increases with the amount of the minority component. However, as discussed in the previous section, the extent of the interchange reactions was higher in the 75/25 blend than in the 90/10 blend. As the copolymers produced should be located in the interphase, they may decrease the interfacial tension of the 75/25 blend more than the mixed component in the 90/10 blend.

In the 50/50 composition [Fig. 7(c)], in spite of the closeness to the phase inversion, the morphology of the dispersed phase is still homogeneous and comparatively small. As can also be seen, the matrix of this intermediate composition was PTT. This agrees with the higher viscosity of phenoxy, when compared with that of PTT at the processing temperature, as the less viscous PTT tends to encapsulate the more viscous phenoxy. The holes in Figure 7(c) do
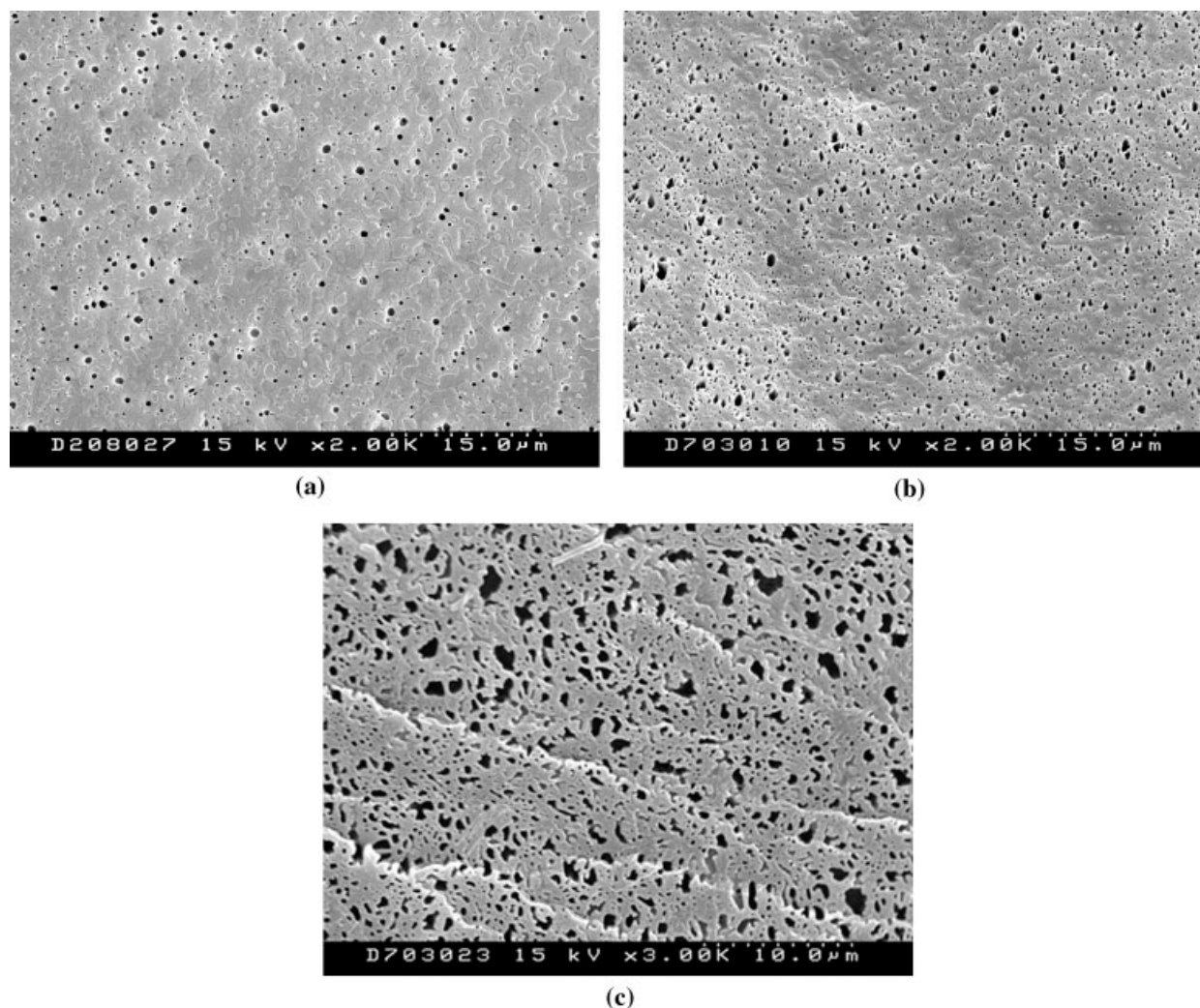
Figure 7 SEM micrographs of the etched surfaces of cryofractured tensile specimens of PTT/phenoxy blends at phenoxy contents of (a) 10%, (b) 25%, and (c) 50%.

not take up half the surface, as the composition indicates. This is attributed to the partially crystalline nature of PTT and to a possible additional PTT crystallization in the presence of chloroform. Solvent-induced crystallization takes place in PTT in the presence of methylene chloride.[21] The crystalline phase should hinder the solvent penetration and dissolution of phenoxy and lead to a smaller hole content.

Mechanical properties

Figure 8 shows the Young's modulus of the blends as a function of composition. As can be seen, a synergistic behavior was found for all compositions. The deviation was small in the 90/10 and 10/90 blends, but it was ~ 5% in the rest of the blends. A similar behavior has been reported for other phenoxy blends, which are miscible,[13] partially miscible,[45] and immiscible.[27] This behavior should be a

consequence of (a) a higher crystallinity of PTT in the blends,[46] (b) a densification of the amorphous phase of the blends due to specific interactions between their components,[47] (c) a higher orientation in the blends than in the neat state,[48] and (d) the
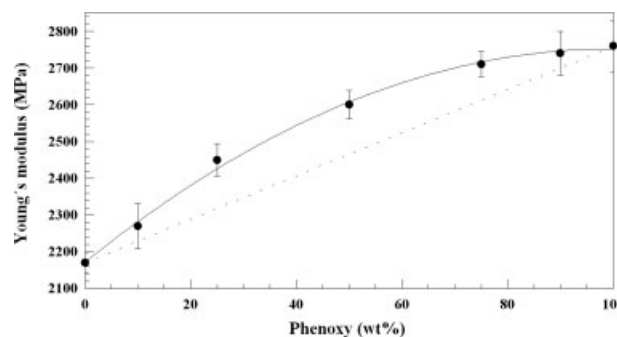

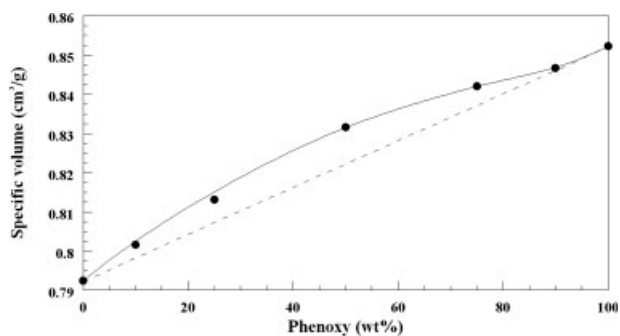**Figure 8** Young's modulus of the PTT/phenoxy blends versus the phenoxy content.

Figure 9 Specific volume of the amorphous part of the blends versus the phenoxy content. The values predicted by the direct rule of mixtures are shown as a broken line.

Figure 11 Yield stress of PTT/phenoxy blends versus phenoxy content.

presence of grafted/crosslinked copolymers, stiffer than the linear homopolymer chains, as a product of interchange reactions.

Interchange reactions between PTT and phenoxy were only observed in PTT-rich compositions. However, no curvature change in the modulus plot that could suggest the presence of such an effect was seen at intermediate compositions. Therefore, the reactions do not appear to have a significant effect on the modulus.

With respect to a possible mixing-induced loss of free volume in the amorphous phases, the specific volume of the amorphous part of the blends was calculated from the experimental density values of the whole blends and the crystallinity content, and it is shown against composition in Figure 9. As can be seen, a positive deviation from linearity appeared. The deviation is significant because a specific volume change of 1% (similar to that reported in this work)[34] led to deviations of roughly 6% in the modulus. Therefore, there was a mixing-induced increase in the specific volume, which would lead to lower modulus.

The orientation of the neat components and of the blends was estimated by means of birefringence measurements in a polarized light microscope. As observed in Figure 10, the orientation of the blends
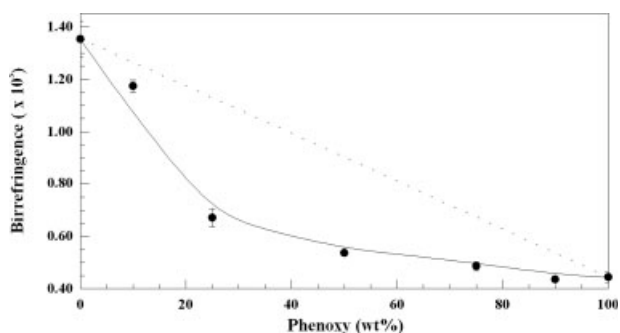
was below that predicted by the direct rule of mixtures. Therefore, it should lead to a modulus decrease. Finally the crystallinity of the blends (Fig. 6) was higher than that of the pure PTT. Moreover, it exhibited a trend with composition similar to that of the modulus, with a maximum positive deviation from linearity of 26% for the 50/50 composition. This agrees with the modulus behavior and should account for the observed modulus increases. Thus, the modulus behavior is attributed to the higher crystallinity of PTT in the blends, compared with the neat state, which overcomes the decrease in the modulus caused by the changes of orientation and specific volume.

Figure 11 shows the yield stress–composition relationship. The trend is similar to that of the Young's modulus at PTT-rich compositions,[49] but in Phenoxy-rich blends there are no positive deviations. Usually the plots of the modulus of elasticity and the yield stress are similar and their shape is attributed to the same structural reasons. However, similar behaviors to that of this study have been seen in other polymer blends.[50]

Figure 12 shows the ductility, measured as the elongation at break, against the blend composition. As can be seen, despite the biphasic structure of the blends, the values are close to those predicted by the
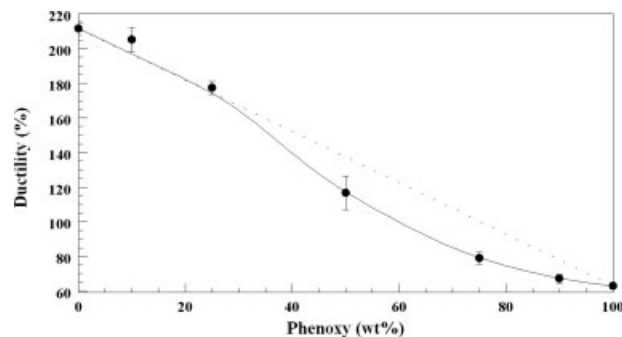

Figure 10 Birefringence of PTT/phenoxy blends versus phenoxy content.

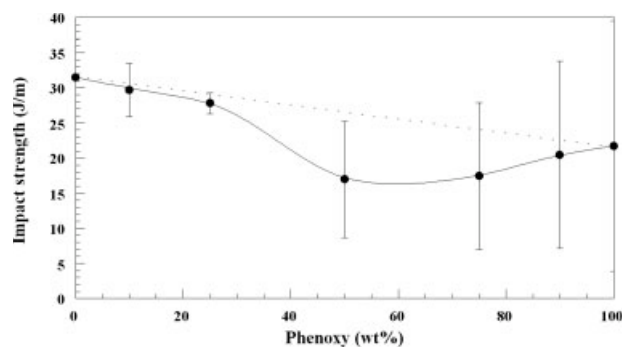**Figure 12** Ductility of PTT/phenoxy blends versus phenoxy content.

Figure 13 Notched impact strength of PTT/phenoxy blends versus phenoxy content.

direct rule of mixtures (linearity). This behavior indicates that the blends are clearly compatible and is attributed to the good interface adhesion between the matrix and the dispersed phase, discussed earlier, and to the small particle size observed in PTT-rich blends. Both a good interphase adhesion[25,51] and a small particle size[25,50,51] have positive effects on the ductility of polymer blends. The values of the phenoxy-rich blends have to be emphasized because though interchange reactions did not occur the blends remained ductile. This positive behavior could be predicted from the cohesive fracture observed in the morphology section.

Finally, the notched impact strength is shown in Figure 13. The values of the neat components are very low due to their high notch sensitivity. The behavior of the blends follows the same trend as that of the elongation at break and is attributed to the same structural parameters. The negative deviations for intermediate and phenoxy-rich compositions are more notable than those of the ductility. This is because a higher adhesion is important in the slow strain–stress tests. In the fast and located fracture of the impact tests, however, the particle size and distribution have the most fundamental role.

CONCLUSIONS

PTT/phenoxy blends are composed by a crystalline PTT phase and two amorphous phases rich in PTT and phenoxy, in which minor amounts of the other component are present. In PTT-rich blends, the minor component is present partially as reacted copolymer. The joint analysis of the miscibility level of the PTT/phenoxy blends of this work with those of the PBT/phenoxy, PET/phenoxy blends, and other blends with polyalkylene terephthalates in the literature, indicates that a miscibility limit exists in blends based on polyalkylene terephthalates with other polymers such as phenoxy, PEI, and PAr. Blends with methylene/carbonyl ratio less than 3/1 in the diol component are miscible.

Blending induced a higher crystallinity of PTT in the blends than in the unblended state, a positive volume of mixing and a smaller orientation in the blends than in the neat state. The observed positive deviations of the modulus of elasticity from the values predicted by the rule of mixtures, and those of the yield stress in PTT-rich blends, are attributed to the combined influence of these three structural characteristics. The approximately linear behavior with composition of the ductility, and to a minor extent of the impact strength, were attributed both to the joint effect of the fine particle size of PTT-rich blends and to the significant interfacial adhesion. Adhesion is attributed to the partial miscibility and was revealed by the cohesive fracture observed whatever the composition.

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