Phase Behavior of Ternary PBT–PC/Phenoxy Blends

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SYNOPSIS

Ternary polymer blends were obtained by melt mixing, mixing up to 30% poly(butylene terephthalate) (PBT) with polycarbonate (PC) and phenoxy in an attempt to improve the miscibility of the PC/phenoxy binary blend. Although most of the blends with a PBT content higher than 10% appear as transparent, two T_g's appeared at all the blend compositions. These T_g's correspond to PC-rich and phenoxy-rich phases where a low amount of the main component of the other phase and all PBT are dissolved in amounts that are a function of the PC/phenoxy ratio of the blend. Increasing the PBT contents in the blends closes to linearity the torque versus composition plot, so that a relationship between miscibility level and viscosity exists in these blends.

INTRODUCTION

The development of new materials based on polymer blends is becoming one of the most active areas of plastics research thanks to the increasing need for new polymers with improved properties for many different applications. Among polymer blends, binary blends are to date the most often studied and commercialized. Many polymer blends have rubbery materials as a second component, but great attention has also been paid to blends whose components are polyesters or other engineering materials.

It is known that miscibility level plays a definitive role in the solid state and mechanical properties of polymers. As a consequence, various attempts have been made to improve the miscibility level of immiscible or partially miscible polymer blends. One such attempt is to add a third component, miscible[1-8] or at least partially miscible[9-11] with the two components of the immiscible blend. This method usually occasions more difficulties in control and optimization of processing and composition of polymer blends, but it seems the natural direction for polymer research to take as seen from the historical development of other materials.

Among condensation polymers both bisphenol A polycarbonate (PC)[12-25] and poly(hydroxy ether of

bisphenol A) (phenoxy)[26-33] have been widely studied as components of binary polymer blends. However, despite the potential miscibility of phenoxy with polyesters or related materials, the PC/phenoxy blend is not fully miscible,[34] although a favorable balance of properties can be produced in this blend through interchange reactions in the melt state.[35,36]

If we look at the possibility of combining PC/phenoxy blends with a third component, we realize, first, that PBT is fully miscible with phenoxy, second, that a partially miscible PC/PBT blend with a PBT content of 44%[13] (Xenoy CL General Electric) has been commercialized, and, finally, that studies have been done of the binary blends PC/PBT,[12-20,33,37,38] PBT/phenoxy,[26,33] and PC/phenoxy.[34-36] Moreover, PC, in spite of its high ductility, stability, and outstanding impact resistance, has a high melt viscosity and poor hydrolysis resistance. PBT is unstable at high temperatures[26,35] but has excellent solvent resistance and high processability; it is also cheaper than the binary PC/phenoxy blend. Such facts render the study of the ternary PBT–PC/phenoxy blends clearly important from a scientific and technological point of view.

For the above reasons, in this study PBT–PC/phenoxy blends with PBT content of 0, 10, 20, and 30% have been obtained by melt mixing in order to determine the ability of PBT to improve the compatibility level of PC/phenoxy blends. Moreover, the change of the plots of the torque of blending

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versus composition when the PBT content increases, that is, when miscibility probably increases, permits testing for the existence of a relationship between miscibility level and torque (viscosity) behavior in these blends.

EXPERIMENTAL

The polymers used were commercial polycarbonate (PC) (Makrolon 2805, Bayer), poly(hydroxy ether of bisphenol A) (phenoxy) (PKHH, Union Carbide), and poly(butylene terephthalate) (PBT) (Pocan 1505, Bayer).

Pellets of the three polymers were dried for a minimum of 48 h, in air at 110°C in the case of PC and PBT, and *in vacuo* at 70°C in the case of phenoxy. After conditioning, they were mixed to the desired PBT–PC/phenoxy compositions.

The PC/phenoxy compositions studied were 0/100, 20/80, 40/60, 60/40, 80/20, and 100/0. These PC/phenoxy proportions were maintained after the addition of 10, 20, and 30% PBT to the blend. With the aim of providing a quick reference to the PC/phenoxy content of the ternary blends, the first number will indicate the PBT content and the two following numbers the PC/phenoxy content of the blend considered without PBT. So that a 10–80/20 blend has a real PBT/PC/phenoxy content of 10/72/18.

Melt blending was carried out in a Brabender Plasticorder equipped with a mixing head at 240°C and 30 rpm following the procedure used with other previous blends.[39] Torque–time plots were recorded by the Brabender. The torque decreases very quickly when the materials melt, until, after a blending time of approximately 11 min, it reaches a steady value. The blends that correspond to this steady value were those used in this work. They appeared to be homogeneous in a macroscopic sense and corresponded to the minimum processing time available for blending.

The blends were processed by compression at 240°C into sheets ($\simeq$ 150 × 150 × 1 mm) and quenched in chilled water. The compression sheets were protected during cooling by two aluminum films adjacent to the polymer during processing, and thereafter protected against moisture absorption in a desiccator. This cautious thermal treatment, due to the presence of PBT, the slight compression, and the different conditioning of the pellets, gave rise to different physical properties in the PC/phenoxy blends of this study as compared with those of previous work.[35] For the sake of comparison, binary

PC/phenoxy blends, too, were obtained at the stated conditions.

Specimens for dynamic mechanical tests and for DSC were obtained from the compression-molded sheets. The dynamic mechanical tests were carried out in a DMTA (Polymer Laboratories), conducted in a single cantilever mode at 1 Hz and 4°/min. After cooling of the specimens, a second scan was performed at the same conditions. DMTA results on other ternary compositions than those stated above were also obtained from blends with a PBT content of 10 and 20%, processed in a similar way, and tested at the same conditions. DSC measurements were carried out in a Perkin-Elmer DSC-2 calorimeter equipped with a Perkin-Elmer TADS system at a scan rate of 20°/min. After the first scan, the sample was quickly cooled, and a second scan was performed at the same conditions.

The determination of the miscibility limits by means of the values of the interaction parameters calculated from the melting point decrease of PBT could not be done due to the presence of kinetic effects together with the thermodynamic ones. This prevents the calculation of a meaningful interaction parameter in this case.

RESULTS AND DISCUSSION

Transparency

A prospective study of miscibility was carried out by the observation of the transparency of the blends in the solid state. With respect to the miscibility level of the binary blends, it is known that PBT/phenoxy blends are fully miscible[26] and that both PBT/PC and PC/phenoxy show partial miscibility,[34,37] i.e., each phase in the binary blends contains an appreciable amount of the other component. This amount is lower in the case of PC/phenoxy blends than in the case of PBT/PC blends.

As was expected, all the PC/phenoxy blends were opaque but the opacity range clearly decreased as the PBT content increased. In fact, the 10–10/90 and 10–90/10 blends appeared as transparent as did all the blends with a 20 or 30% PBT content, except the binary PBT/PC blends. This testifies to the existence of miscibility in most of the blends with either 20 or 30% PBT. However, it must be taken into account that the difference of index of refraction between PC (1.585)[40] and phenoxy (1.598)[28] is close to that which can produce transparency in spite of immiscibility ($\Delta n \times 10^2 \leq 0.5$–1).[41] In addition to this, in this ternary blend the probability of trans-

parency due to the similarity of indices of refraction may be enhanced by the confluence of the indices of refraction of the two amorphous phases as a result of the presence of PBT, so that the observed transparency in this case, instead of being an indication of miscibility, may be a result of the confluence of the indices of refraction.

The levels of crystallinity of the ternary blends before the first scan, measured by DSC, are shown in Table I. These levels of crystallinity were calculated from the difference between the melting and crystallization enthalpies observed in the scans. In the case of the crystallinity level of the 10% PBT blends mainly, the reliability of the results must be conditioned by the experimental error which is comparable to the obtained crystallinity values. An enthalpy of fusion of 31.80 kJ/(mol of monomer unit) was taken for the 100% cristalline PBT.[42] Two crystallization exotherms were observed in most of the ternary blends, mainly in those with high PBT contents. As can be seen, these crystallinity levels, even taking into account factors like recrystallization during heating,[2,43,44] seem to be too high to be compatible with the transparency observed in most of the blends with 20 or 30% PBT content. However, this appearance of transparency in rather crystalline blends or polymers, although surprising, should be considered more common than before, because it has also been observed in partially crystalline PBT/ phenoxy blends[26] and in PBT/PC blends.[37]

Dynamic Mechanical Properties and Miscibility State of the Blends

In Figure 1 appear the dynamic mechanical spectra corresponding to the 40/60 PC/phenoxy blends, with 0% PBT (first scan) as a reference [Fig. 1(a)], and with 10, 20, and 30% PBT [Figs. 1(b), 1(c), and 1(d)] (first and second scans). As can be seen,

Table I PBT Crystallinity Level of Several Blends by DSC

Blend Composition (PBT-PC/Phenoxy)	Crystallinity Level of the Blends (%)
10–80/20	2.3
10–100/0	1.5
20–80/20	3.4
20–100/0	4.7
30–80/20	6.9
30–100/0	6.9
100–0/0	32.0

and as happened with the other studied blends, the blends containing PBT show two T_g's which change with PBT content in both of the scans. The 0–40/ 60 blend of Figure 1(a) shows a single T_g because the T_g of the PC-rich phase could not be observed due to the too-soft nature of the matrix. This demonstrates the presence of more than one amorphous phase in the blends and the incomplete miscibility of the blends. No change of moduli or tan δ was seen in the zone where the T_g of PBT should have appeared, so that all PBT, at these PBT contents, must have been present either in the PC or in the phenoxy phases. The small peaks in the tan δ plot which appear before the T_g of PC and at $\simeq$ 40°C above the T_g of phenoxy in the first scan correspond to PBT crystallization, as they appear together with a modulus increase, clearly decrease in the second scan, do not appear in the PC/phenoxy blend, and are greater when the PBT content of the blend increases. This crystallization during the first scan is also indicated by the higher storage modulus in the second scan in all the temperature range than in the first scan.

Figure 1 shows the main secondary transitions of both PC and phenoxy in the −150−−50°C temperature range. The origins of these transitions have previously been studied.[26,35,37,45] The shape of the secondary transitions of the ternary blends was that of the principal component and the intensity of the transitions was nearly constant. Similar behavior appeared in the dynamic plots of the other studied blends. That is, PBT presence does not show any remarkable negative influence on these low-temperature secondary transitions which are so related to the most important mechanical properties of both polycarbonate and phenoxy.
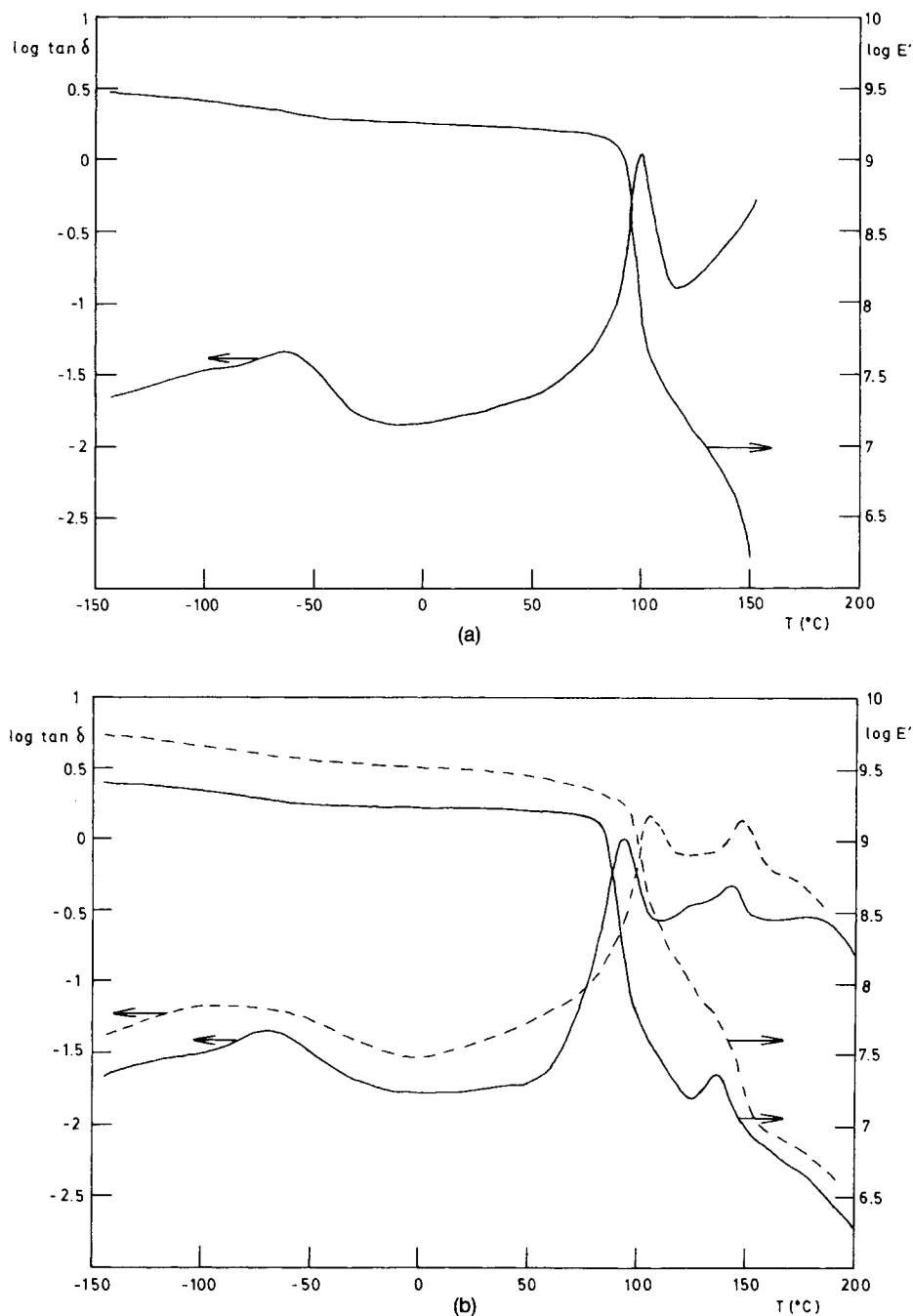
That is, in spite of (a) the miscibility and partial miscibility of PBT with phenoxy and PC respectively, (b) the transparency of most of the blends with high PBT content, and (c) the distribution of all the added PBT in the PC and phenoxy phases, a distribution which must improve the miscibility of the blend, the studied PBT-PC/phenoxy blends of low PBT content are not fully miscible. However, PBT contents higher than 30% may well give rise to miscibility, as was reported for other similar ternary blends, one with poly(ϵ-caprolactone) instead of PBT as third component[1] and the other with polyarylate instead of PC.[2]

Phase Behavior

The variations of the T_g's with composition when PBT is added to PC/phenoxy blends, and which

Figure 1   Dynamic mechanical spectra of (a) 0–40/60, (b) 10–40/60, (c) 20–40/60, and
(d) 30–40/60 PBT-PC/phenoxy blends during the first (——) and second (— — —) scans.
Second scan plots are moved 0.25 units upward.

were obtained from the tan δ peak data, are collected
in Figure 2 for the first scan [Fig. 2(a)] and the
second scan [Fig. 2(b)] against PC/phenoxy com-
position. T_g measurements of the PC phase of the
PC/phenoxy blends with lower PC content than
60% could not be made because of the too-soft nature

of the phenoxy matrix of the blends at these tem-
peratures.

With respect to the differences between the two
scans, most of the PBT blends, transparent or al-
most transparent before the first scan, became com-
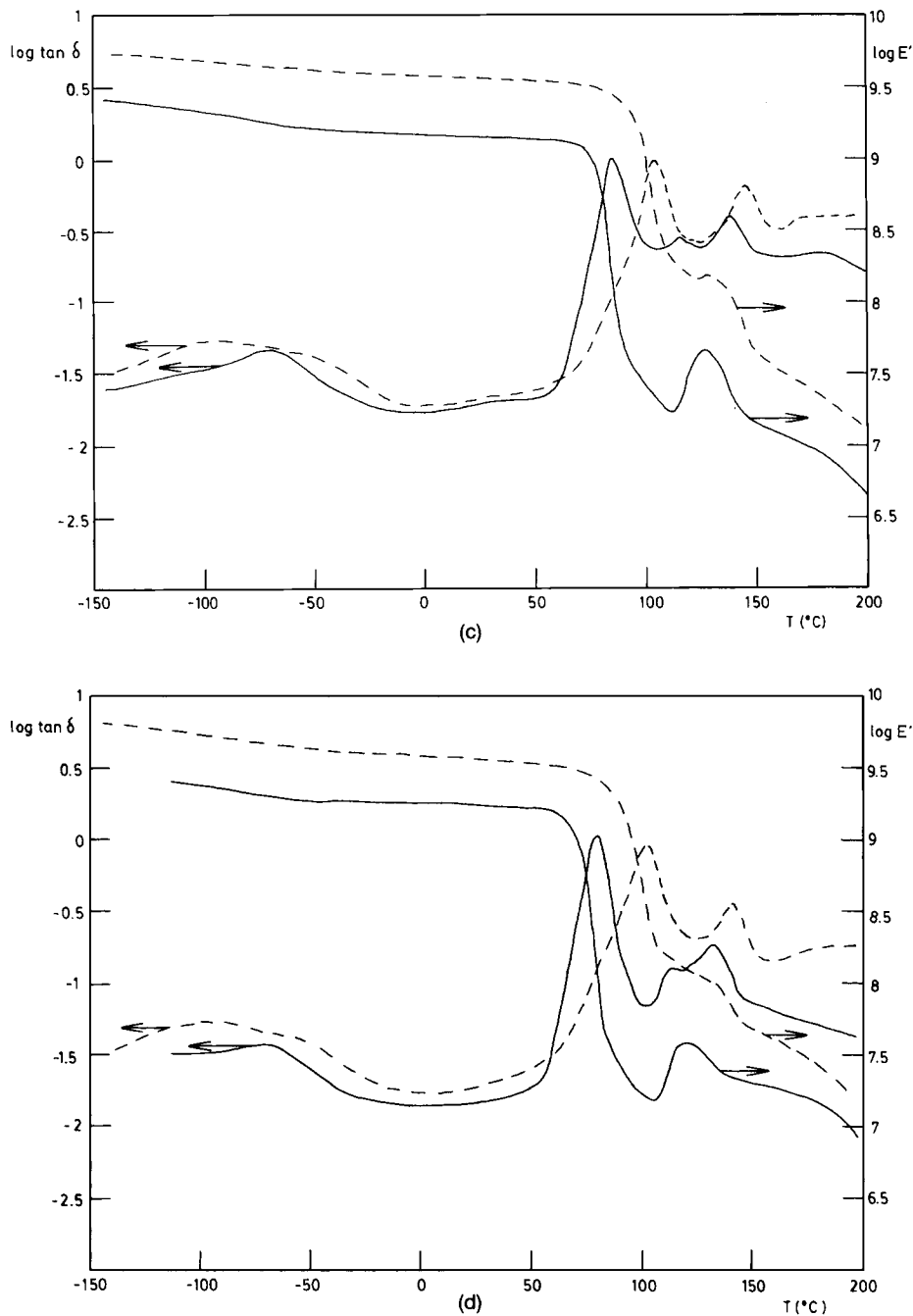pletely opaque after the first scan. This opacity must

Figure 1 (*Continued from the previous page*)

be due to the crystallization of amorphous PBT during the first scan. As a result, this posterior crystallization will produce a decrease of the amorphous PBT content in the material; as a further consequence, the second scan will inform us about the phase behavior of blends with a different amorphous/crystalline PBT relation.

As can be seen in Figure 2(a), the two T_g's of the

PC/phenoxy binary blends are not those of the pure polymers. This proves the presence of phenoxy in the PC phase and of PC in the phenoxy phase. As can also be seen in Figure 2(a), the presence of increased PBT contents lowers the T_g of both the PC and phenoxy phases. This again proves the presence of PBT in both phases of the blend. Thus, the ternary blends must be composed of a PC-rich phase
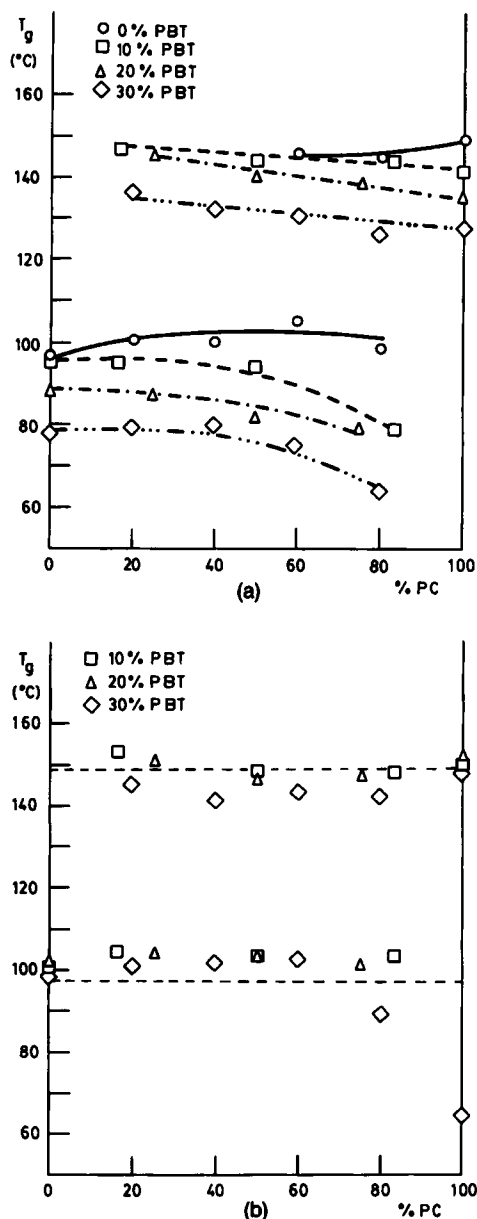
Figure 2 T_g vs. PC content of PBT–PC/phenoxy blends as measured in the first (a) and second (b) DMTA scans for different PBT contents. The reference lines drawn in (b) correspond to the T_g's of pure PC and phenoxy.

with small amounts of PBT and phenoxy and a phenoxy-rich phase with small amounts of PBT and PC.

If we look at the dependence of the T_g of the PC-rich phase of the ternary blends on composition, we realize that when phenoxy is progressively substituted for PC in PC-rich blends, i.e., when phenoxy content increases, the T_g of the PC-rich phase increases. The T_g of the PC-rich phase should have decreased due to the probable increased presence of

phenoxy in the PC phase at increasing phenoxy contents of the blend. This failure of the T_g to decrease testifies to a great decrease of the PBT amount in the PC-rich phase and to PBT increasing presence in the phenoxy-rich phase when phenoxy content of the blend increases. This indicates a greater affinity of PBT for the phenoxy-rich phase than for the PC-rich phase when both phases are together. This greater affinity is in accord with the greater miscibility of PBT with phenoxy[26] as compared to that with PC[37] and must give rise to a higher presence of PBT in the phenoxy-rich phase than in the PC-rich phase. This higher presence of PBT in the phenoxy phase also causes the greater T_g decrease of the phenoxy-rich phase as related to the corresponding T_g decrease of the PC-rich phase, both of the decreases taking as a reference the corresponding T_g–composition plots of the binary PC/phenoxy blends.

As can also be seen in Figure 2(a), the T_g's of the phenoxy-rich phase of the PC/phenoxy blends show the usual behavior of binary blends; that is, they change towards the T_g of the other component of the blend. However, the behavior of the ternary blends is again surprising because, at increasing phenoxy contents, the T_g's of the phenoxy-rich phases increase at all the PBT contents. This is in spite of (a) the probable decrease of the PC content of the phenoxy-rich phase at increasing phenoxy contents and (b) the increasing total amount of PBT in the phenoxy-rich phase (because of the above-mentioned relative and absolute decrease of PBT content of the PC-rich phase).

This increase of the T_g's of the phenoxy-rich phase at increasing phenoxy contents must be due to the fact that the decrease of PBT content in the PC-rich phase at decreasing PC contents is lower than that which the increased phenoxy content needs to maintain the composition of the phenoxy-rich phase. This is due to the greater PBT presence in the phenoxy phase than in the PC phase in these ternary blends, which in turn is due to the higher affinity of PBT for phenoxy than for PC. This explanation gives as a result a decrease in the relative, but not the absolute, PBT content of the phenoxy-rich phase. Thus, increased phenoxy contents in the ternary blends give rise to a decrease of the relative amount of PBT in both phases due to the substitution of the high-PBT-content phenoxy phase for the low-PBT-content PC phase.

In Figure 2(b), the increased mobility restrictions, due to the increased crystalline PBT content in both phases during the second scan, bring about higher T_g's of both the PC and phenoxy phases of

the ternary blends observed in Figure 2(b) as compared to those of Figure 2(a). These T_g increases were expected from the crystallization peaks observed by DMTA in the first scan and show the expected overall influence of the decrease of amorphous PBT contents on the T_g's of the PBT-PC/phenoxy blends. The observed change appears to be stronger in the phenoxy-rich phase where the T_g's, rather different from those of phenoxy in the first scan, showed a significant tendency to approach the T_g of phenoxy in the second scan. This indicates that PBT crystallization is easier from the phenoxy phase than from the PC phase in the stated conditions.

The crystallization (T_c) and melting temperatures (T_m) corresponding to the first DSC scan of several blends are collected in Table II. As can be seen, the T_m values are fairly low as compared with that obtained for pure PBT (226°C). This is an indication of the existence, as mentioned above, of kinetic effects superimposed on the thermodynamic ones.

The crystallization temperatures increase as the PBT content of the blend decreases. This is true for the high-temperature crystallization peak of the PC phase and also for the low-temperature, fairly sharp peak of the phenoxy phase. As has been suggested,[46] it seems that this is due to the kinetic hindrance of PBT crystallization caused by the presence of other soluble components.

Finally, if we look at the T_c from the phenoxy-rich phase in Table II and at the corresponding T_g's in Figure 2(a) (80/20 PC/phenoxy blend with different PBT contents), we realize that the higher the T_g of the phenoxy phase is, the greater the T_c appears. The same effect appears in the case of the T_c's from the PC-rich phase. That is, it seems that, as has been indicated,[2] the T_c values are related to those of the T_g. Trends of T_c versus composition similar to those of T_g were also observed in other blends.[2,23,46,47]

Torque-Composition Relation

As is known, several studies and contradictory propositions have been done to find out if there exists a relationship between miscibility level and the behavior of the plot of either viscosity or torque of blending versus composition. Some of these works indicate that miscibility gives rise to a plot close to linearity, although immiscibility would give rise to plots far from linearity[48-52]; however, other works indicate that such a relationship does not exist.[53]

The work reported herein, which produced PC/phenoxy blends with an increased amount of PBT, offers a valuable opportunity of finding out which of the above propositions is true in this case, because increasing PBT contents gives rise to an overall increase in miscibility. For these reasons, Figure 3 shows blending torque corresponding to the minimum torque, after a blending time of approximately 11 min, versus composition plots for the four different PBT-PC/phenoxy blends.

As can very clearly be seen, the higher the PBT content is, that is, the higher miscibility is, the closer the torque-composition curve appears to linearity. Thus, at least in this case, a relationship between miscibility level and behavior of the torque-composition curve does exist.

CONCLUSIONS

Whatever the PC content of the binary blend, PBT contents up to at least 30% completely distribute in the two phases of PC/phenoxy blends, when melt mixed together. This improves the miscibility of the PC/phenoxy blends. However, despite the overall transparency increase observed when PBT content increases, all the blends show two clear T_g's at all the PC/phenoxy blend contents and at all the studied PBT contents.

Table II   Crystallization (T_c) and Melting (T_m) Temperatures of PBT Corresponding to the First Scan in the DSC

Blend Composition (PBT-PC/Phenoxy)	T_c from Phenoxy-Rich Phase (°C)	T_c from PC-Rich Phase (°C)	T_m (°C)
10–100/0	—	186	221
20–100/0	—	182	223
30–100/0	—	178	223
10–80/20	133	—	217
20–80/20	108	194	218
30–80/20	101	180	220

Torque
(N·m)

○   0% PBT
□ 10% PBT
△ 20% PBT
◇ 30% PBT
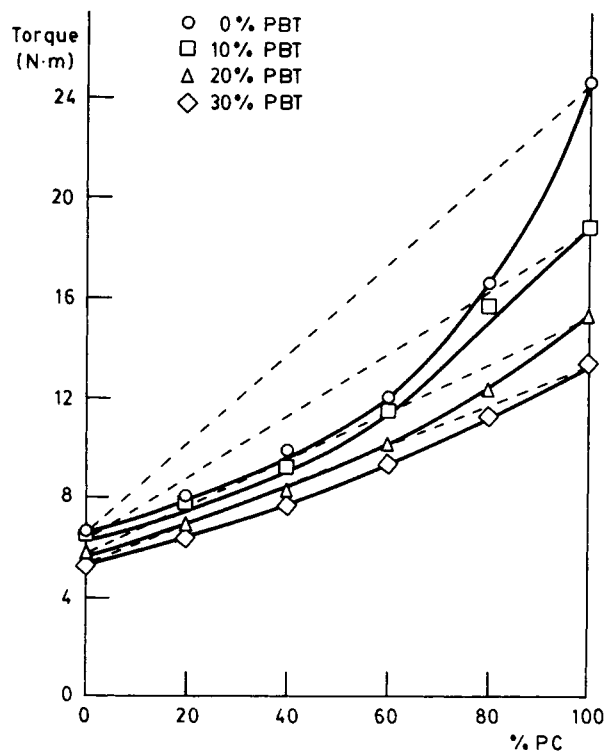
Figure 3   Torque vs. PC content of PBT–PC/phenoxy blends with different PBT contents. The discontinuous straight lines correspond to the additivity law. PC contents are those of the assumed binary PC/phenoxy blends.

The T_g-composition plot of this ternary blend shows unusual behavior because the two T_g's, instead of approaching one another, separate from each other. This is due to the different amount of PBT in each phase when the PC/phenoxy relation changes. Moreover, increased phenoxy contents in the ternary blends give rise to a decrease in the relative amount of PBT in both phases. This is due to the substitution of low PBT content PC phase by the high PBT content phenoxy phase. The absolute amount of PBT in the phenoxy-rich phase, however, increases.

PBT presence does not show any remarkable low-temperature suppression effect on the secondary transition of either PC or phenoxy; this indicates that the remarkable mechanical properties of these polymers caused by these transitions will probably not change.

In this ternary biphasic blend there exists a relation between miscibility level and torque/viscosity behavior because an increase of PBT content, that is, a miscibility increase, gives rise to a progressive approach of the torque–composition curves to linearity.

REFERENCES

1. W. H. Christiansen, D. R. Paul, and J. W. Barlow, *J. Appl. Polym. Sci.,* **34,** 537 (1987).
2. J. I. Eguiazabal, J. J. Iruin, M. Cortazar, and G. M. Guzman, *J. Appl. Polym. Sci.,* **32,** 5945 (1986).
3. T. K. Kwei, H. L. Frisch, W. Radigan, and S. Vogel, *Macromolecules,* **10,** 157 (1977).
4. B. Ameduri and R. E. Prud'Homme, *Polymer,* **29,** 1052 (1988).
5. J. J. Iruin, J. I. Eguiazabal, and G. M. Guzman, *Eur. Polym. Sci.,* **25,** 1169 (1989).
6. S. H. Goh, K. S. Siow, and K. S. Yap, *Thermochim. Acta,* **102,** 281 (1986).
7. S. H. Goh and K. S. Siow, *Thermochim. Acta,* **105,** 191 (1986).
8. W. W. Y. Lau, T. Swee-Hin, and G. Suat-Hong, *Br. Polym. J.,* **20,** 323 (1988).
9. V. S. Shah, J. D. Keitz, D. R. Paul, and J. W. Barlow, *J. Appl. Polym. Sci.,* **32,** 3863 (1986).
10. J. C. Huarng, K. Mim, and J. L. White, *Polym. Eng. Sci.,* **28,** 1085 (1988).
11. Y. Wang and S. Chen, *Polym. Eng. Sci.,* **21,** 47 (1981).
12. J. Devaux, P. Godard, and J. P. Mercier, *Polym. Eng. Sci.,* **22,** 229 (1982).
13. A. W. Birley and X. Y. Chen, *Br. Polym. J.,* **16,** 77 (1984).
14. A. W. Birley and X. Y. Chen, *Br. Polym. J.,* **17,** 297 (1985).
15. J. Devaux, P. Godard, and J. P. Mercier, *J. Polym. Sci. Polym. Phys. Ed.,* **20,** 1875 (1982).
16. S. Y. Hobbs, M. E. J. Dekkers, and V. H. Watkins, *J. Mater. Sci.,* **23,** 1219 (1988).
17. M. E. J. Dekkers, S. Y. Hobbs, and V. H. Watkins, *J. Mater. Sci.,* **23,** 1225 (1988).
18. S. Y. Hobbs, V. L. Groshhans, M. E. J. Dekkers, and R. A. Shultz, *Polym. Bull.,* **17,** 335 (1987).
19. S. Y. Hobbs, M. E. J. Dekkers, and V. H. Watkins, *Polym. Bull.,* **17,** 341 (1987).
20. D. Delimoy, C. Bailly, J. Devaux, and R. Legras, *Polym. Eng. Sci.,* **28,** 104 (1988).
21. M. Gaztelumendi, I. Mondragon, and J. Nazabal, *Makromol. Chem. Macromol. Symp.,* **20/21,** 269 (1988).
22. A. Golovoy, M. F. Cheung, and H. Van Oene, *Polym. Eng. Sci.,* **27,** 1642 (1987).
23. E. A. Joseph, M. D. Lorenz, J. W. Barlow, and D. R. Paul, *Polymer,* **23,** 112 (1982).
24. P. Godard, J. M. Dekoninck, V. Devlesaver, and J. Devaux, *J. Polym. Sci. Polym. Chem. Ed.,* **24,** 3301 (1986).
25. P. Godard, J. M. Dekoninck, V. Devlesaver, and J.

Devaux, *J. Polym. Sci. Polym. Chem. Ed.*, **24,** 3315 (1986).

26. L. M. Robeson and A. B. Furtek, *J. Appl. Polym. Sci.*, **23,** 645 (1979).

27. J. E. Harris, S. H. Goh, D. R. Paul, and J. W. Barlow, *J. Appl. Polym. Sci.*, **27,** 839 (1982).

28. V. B. Singh and D. J. Walsh, *J. Macromol. Sci. Phys.*, **B25,** 65 (1986).

29. R. W. Seymour and B. E. Zehner, *J. Polym. Sci. Polym. Phys. Ed.*, **18,** 2299 (1980).

30. C. Uriarte, J. I. Eguiazabal, M. Llanos, J. I. Iribarren, and J. J. Iruin, *Macromolecules,* **20,** 3038 (1987).

31. B. T. Swinyard, J. A. Barrie, and D. J. Walsh, *Polym. Commun.*, **28,** 331 (1987).

32. I. Mondragon, P. M. Remiro, and J. Nazabal, *Eur. Polym. J.*, **23,** 125 (1987).

33. J. I. Eguiazabal, M. Cortazar, J. J. Iruin, and G. M. Guzman, *J. Macromol. Sci. Phys.*, **B27,** 19 (1988).

34. I. Mondragon, M. Gaztelumendi, and J. Nazabal, *Polym. Eng. Sci.*, **26,** 1478 (1986).

35. I. Mondragon, M. Gaztelumendi, and J. Nazabal, *Polym. Eng. Sci.*, **28,** 1126 (1988).

36. I. Mondragon and J. Nazabal, *J. Mater. Sci. Lett.*, **6,** 698 (1987).

37. D. C. Wahrmund, D. R. Paul, and J. W. Barlow, *J. Appl. Polym. Sci.*, **22,** 2155 (1978).

38. A. Golovoy, M. F. Cheung, and H. Van Oene, *Polym. Eng. Sci.*, **28,** 200 (1988).

39. I. Mondragon and J. Nazabal, *J. Appl. Polym. Sci.*, **32,** 6191 (1986).

40. D. W. Van Krevelen and P. J. Hoftyzer, *Properties of Polymers: Their Estimation and Correlation with Chemical Structure,* 2nd ed., Elsevier, Amsterdam, 1976, Chap. 10.

41. L. Bohn, in *Polymer Handbook,* 2nd ed., J. Bandrupt and E. H. Immergut, Eds., Wiley-Interscience. New York, 1975, p. III 212.

42. K. Tashiro, Y. Nakai, M. Kobayashi, and H. Tadokoro, *Macromolecules,* **13,** 137 (1980).

43. S. Z. D. Cheng, R. Pan, and B. Wunderlich, *Makromol. Chem.,* **189,** 2443 (1988).

44. P. B. Rim and J. P. Runt, *Macromolecules,* **16,** 762 (1983).

45. C. J. T. Landry and P. M. Henrichs, *Macromolecules,* **22,** 2157 (1989).

46. R. N. Mohn, D. R. Paul, J. W. Barlow, and C. A. Cruz, *J. Appl. Polym. Sci.,* **23,** 575 (1979).

47. M. Kimura, R. S. Porter, and G. Salee, *J. Polym. Sci. Polym. Phys. Ed.,* **21,** 367 (1983).

48. C. M. Roland, *J. Polym. Sci. Polym. Phys. Ed.,* **26,** 839 (1988).

49. C. D. Han, Y. W. Kim, and S. J. Chen, *J. Appl. Polym. Sci.,* **19,** 2831 (1975).

50. J. F. Carley and S. C. Crossan, *Polym. Eng. Sci.,* **21,** 249 (1981).

51. K. M. O'Connor, and E. B. Orler, *Polym. Eng. Sci.,* **28,** 1132 (1988).

52. F. P. La Mantia, and A. Valenza, *Eur. Polym. J.,* **24,** 825 (1988).

53. L. A. Utracki, *Polym. Eng. Sci.,* **28,** 1401 (1988).