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CHARACTERIZATION OF MELT-MIXED POLY(ETHYLENE-2,6-NAPHTHALATE) (PEN)/ POLYCARBONATE (PC) BLENDS

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ABSTRACT

Melt blending of poly(ethylene naphthalate) (PEN) and bisphenol A polycarbonate (PC) was performed without the addition of catalyst in a batch mixer at 290°C at various compositions. All the blends prepared exhibited a biphasic character and had very good mechanical properties, in some cases, even better than those of the respective pure constituents. This behavior was attributed to a copolymer formation in the mesophase, which effectively compatibilizes the system. The formation of a PEN/PC block copolymer was considered to be due to transesterification reactions between PEN and PC and was verified by extraction experiments and examination of the soluble and insoluble fractions by various spectroscopic techniques.

Key Words: Polymer blends; Poly(ethylene naphthalate); Bisphenol A polycarbonate; Reactive blending

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INTRODUCTION

Reactive blending is a versatile and low cost way to produce compatible blends that combine the properties of interest of initially immiscible constituents.[1] This compatibility can be accomplished through interchange reactions during melt mixing between reactive functional groups of properly chosen starting polymers, which lead to the formation of a copolymer that plays the role of the compatibilizer.[2]

Polyesters are a major class of polymers that have been extensively studied in reactive processes. In polyester blends, transesterification reactions (such as alcoholysis, acidolysis, or direct ester interchange), which proceed in molten state, result in the formation of initially block and finally random copolymers.[3]

Although most of the scientific and industrial interest has been focused on poly(ethylene terephthalate) (PET), new packaging opportunities demand performance levels that cannot be reached by it. A solution to these demanding requirements is poly(ethylene-2,6-naphthalate) (PEN), which in the future will probably attract increasing attention. PEN has better mechanical strength; higher heat and chemical resistance; better dimensional, thermal, UV and hydrolytic stability; and improved barrier properties.[4] Numerous studies have also been devoted to blends of PET and PEN, from which one would expect to make potential packaging materials with improved thermal and barrier properties.[5–14]

Polycarbonate (PC) is an amorphous polymer with attractive engineering properties, including high impact strength, low moisture absorption, low combustibility, good dimensional stability, and high light transmittance (up to 88%). Blends of PC with other engineering polyesters [15] and especially with PET [3,16–26] and PBT [26–30] have been extensively studied. The main reaction occurring in molten state is transesterification between PC and the other polyester in the presence of catalyst.[3,17,21,23–26,28,30–32] Catalysis of the transesterification reaction can be obtained either by the residual catalyst present in commercial polyesters [3,21,28,30,31] or by freshly added catalysts.[23–26]

In the present work, we report on the melt-mixing behavior of PEN blends with various percentages of PC and the subsequent characterization of the obtained mixtures using tensile testing, dynamic mechanical analysis (DMA), differential scanning calorimetry (DSC), scanning electron microscopy (SEM), extraction experiments, ^{1}H NMR, Fourier transform infrared (FTIR), as well as Fourier transform Raman (FT-Raman) spectroscopies.

EXPERIMENTAL

Materials and Sample Preparation

PEN was purchased from Goodfellow, while Bisphenol-A PC with $\bar{M}_n = 29{,}000$ was obtained from BDH Chemicals Ltd. They were both dried in a vacuum oven at 120°C for 48 hours to remove sorbed water in order to prevent hydrolysis during their melt processing.

Blends were prepared by melt mixing in a stainless steel homemade mixer previously described.[33] Mixing was carried out under a blanket of argon, and the maximum amount produced per batch was ca. 10 g. Temperature and blending time were varied to obtain improved ultimate tensile properties, i.e., elongation at break ε_b (Table 1). Optimum mixing conditions were determined to be 290°C and a mixing time of 10 min. Blending experiments were repeated at least twice to ensure that the blending procedure was reproducible. Compositions prepared under these conditions were 90/10, 85/15, 75/25, 50/50, 25/75, 15/85 (w/w) PEN/PC.

Films were made by compression molding between Teflon sheets at 290°C followed by quenching to 0°C.

Apparatus and Procedures

Tensile tests were performed according to ASTM D882 at 23°C using a J.J. Tensile Tester type T5001 and film strips with dimensions 3.0 cm × 0.65 cm × 0.025 cm. Data reported was obtained at a crosshead speed of 10 cm min^{-1}.

DSC measurements were carried out in an inert atmosphere using the DSC SP plus equipped with Autocool accessory from Rheometrics Scientific Co. Calibration was carried out with indium standard. Sample weight was 5–10 mg, and the heating rate was 20°C min^{-1}. The samples were heated to 300°C, quenched to −50°C, followed by heating to 300°C.

Table 1. Optimization of Mixing Conditions[a]

Temperature (°C)	Time (min)	σ_y (MPa)	σ_b (MPa)	ε_b (%)
290	7.5	–	52 ± 7	9 ± 1
290	10	66 ± 4	55 ± 6	267 ± 16
320	10	–	52 ± 2	9 ± 15
290	15	55 ± 4	56 ± 11	252 ± 76
290	20	54 ± 8	51 ± 7	171 ± 48

[a] Blend PEN/PC 85/15, quenched to 0°C.

Dynamic mechanical analysis was performed in a Solid State Analyzer RSA II, Rheometrics Scientific Co, at 10 Hz. Specimen dimensions were 3.0 cm × 0.9 cm × 0.02 cm.

Extraction experiments were performed in chloroform at room temperature for 3 days in order to remove the nongrafted polycarbonate.

SEM was carried out on cryofractured specimens, etched or not, using a Jeol model 6320-Field Emission Scanning Electron Microscope at a tilt angle of 30°.

FTIR Spectra Were Obtained Using a Perkin-Elmer 1600 Spectrometer

Raman spectra were recorded with a Bruker FRA-106/S FT-Raman attached to an Equinox 55 FT-IR spectrometer. A near-infrared Nd:YAG laser operating at 1064 nm was focused onto the sample with an average of 100 scans at 4 cm^{-1} resolution.

NMR examination was performed by means of high resolution ^{1}H NMR spectroscopy. Spectra were obtained by an Avance DPX 400 MHz. The samples were dissolved in a 2/1 (v/v) mixture of trifluoroacetic acid and deuterated chloroform.

RESULTS AND DISCUSSION

Melt mixing of PEN with PC at 85/15 composition was performed at various temperatures and mixing times in order to evaluate the possibility of reactive blending, and the results are summarized in Table 1. Tensile properties, i.e., tensile strength σ_b and ultimate elongation ε_b, were used as criteria to determine the optimum mixing conditions at which the compatibilization is more effective, since large deformation may be attributed to good component adhesion in blends. As it was expected in this type of mixer, short mixing times (e.g., 7.5 min) do not allow reactions between the two polymers, and as a result very low values of elongation at break are obtained.

Although the mechanical properties of the 85/15 blends prepared at 10 and 15 min do not show significant differentiation, the shorter time was adopted, since a longer reaction time, as well as higher temperature, may contribute to partial decomposition of the constituents of the polymeric blend. This is something that was observed in the case of the blends prepared at 290°C for 20 min and at 320°C for 10 min where mechanical properties are obviously deteriorated. So, optimum mixing conditions for the specific system were determined to be 290°C and a mixing time of 10 min.

Mechanical Properties

Tensile properties in terms of σ_b, ε_b, and yield stress (σ_y) of the blends prepared under optimum mixing conditions in the whole composition range,

Table 2. Ultimate Tensile Properties of Blends, σ_b, ε_b, and σ_y of Quenched Blends Prepared at Optimum Conditions (10 min, 290°C) and Measured at R.T.

PEN/PC	σ_y (MPa)	σ_b (MPa)	ε_b (%)
100/0	–	49 ± 7	8 ± 1
85/15	66 ± 4	55 ± 6	267 ± 16
75/25	56 ± 6	45 ± 7	143 ± 38
75/25[a]	58 ± 2	53 ± 5	194 ± 72
50/50	52 ± 1	41 ± 2	42 ± 1
25/75	46 ± 2	45 ± 4	88 ± 34
15/85	43 ± 3	40 ± 3	49 ± 8
0/100	38 ± 1	40 ± 2	91 ± 17

[a] After 20 days.

as well as of the pure constituents are given in Table 2. The results are the average of five to eight tests. The composition dependence of σ_b, ε_b, and energy to tensile failure E_b (determined as the area under the stress-strain curve) are shown in Figs. 1 and 2, respectively.

The σ_b values of the blends remain constant at the σ_b level of the two components, while ultimate elongation ε_b is improved with the addition of PC in PEN-rich blends. Moreover, all the blends present yielding points. For the PEN-rich blends, σ_y values are higher than the σ_b of pure PEN, while for the PC-rich blends σ_y values are higher than the σ_b of pure PC. It seems that the incorporation of PC in the PEN matrix changes its mode of failure from brittle fracture to a ductile or yielding one. As shown in Fig. 1, the 85/15 blend shows a higher strength value, indicating a synergistic effect. According to Mishra and Deopura,[34] this can be attributed to higher chain entanglement leading to higher crystalline links. This synergism is also observed in the E_b values of the blends of these compositions, as shown in Fig. 2. The intermediate composition (50/50) shows a minimum in the E_b value that may be due to matrix inversion. In general, overall ultimate tensile properties and E_b values are good and in most cases even enhanced compared to those of the pure constituents. The examination of the tensile properties of the 75/25 blend after 20 days showed that no effects due to physical aging were observed (Table 2).

The dynamic mechanical behavior of the PEN/PC blends is shown in Figs. 3 and 4 in terms of the temperature dependence of the storage (E′) and loss (E″) modulus. The temperature dependence of the storage modulus E′ (Fig. 3) reveals that a two-phase system is obtained. The stiffness of the blends though, is maintained at high levels, similar to this of PEN, despite of the incorporation of PC. As we can see in the temperature dependence plots of the loss modulus E″ (Fig. 4), the main relaxation of PEN associated with T_g is located at ca. 128.6°C (α), while the secondary subambient temperature relaxation is detected at 61.5°C. PC shows a T_g at 156°C. The secondary

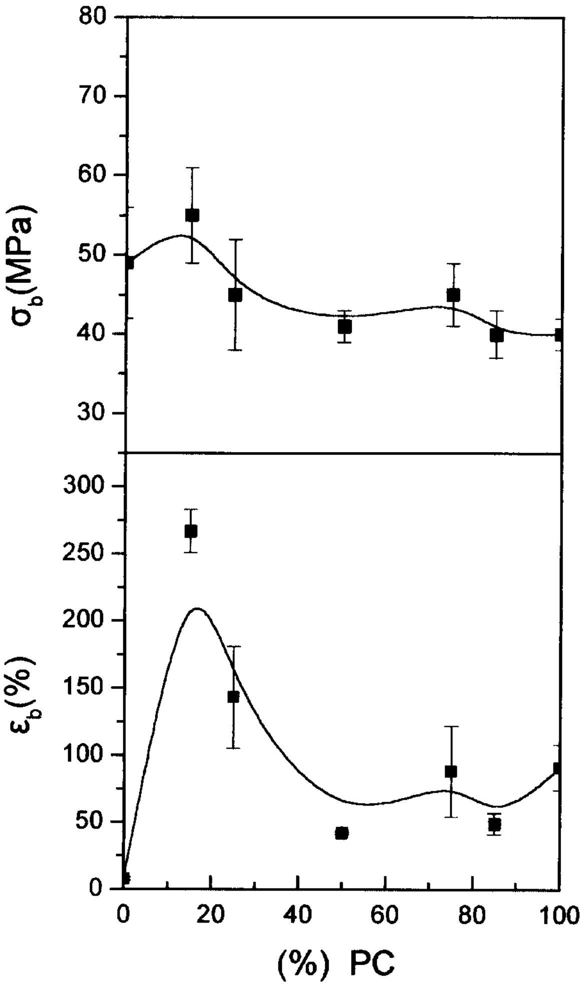


Figure 1. Composition dependence of ultimate properties of PEN/PC blends.

relaxation of PEN does not show any noticeable shift in its blends with PC. Although the T_g's of PEN and PC seem to shift towards each other with the composition (Table 3) (up to 1.4°C for PEN and 5°C for PC), these shifts are not so profound to be accounted.

The presence of two phases combined with the very good mechanical properties described above supports the view that a very effective compatibilization has been achieved in the studied system. Such a compatibilization is attributed to the presence of a copolymer formed by interchange reactions that may occur during the melt mixing of the two polyesters. Such reactions lead to the *in situ* formation of copolymers at the interface leading in higher compatibilization efficiency.[3] This assumption was further evaluated and the results are stated below.

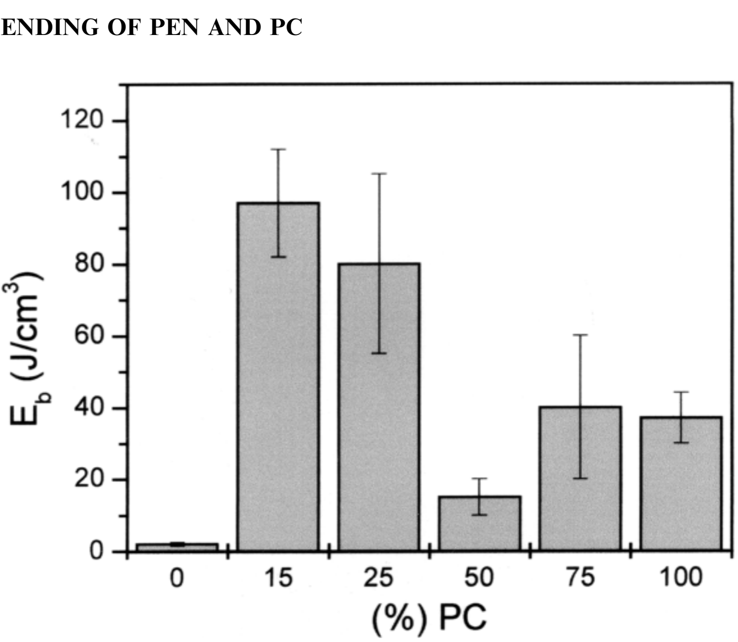


Figure 2. Composition dependence of energy to tensile failure E_b of PEN/PC blends.

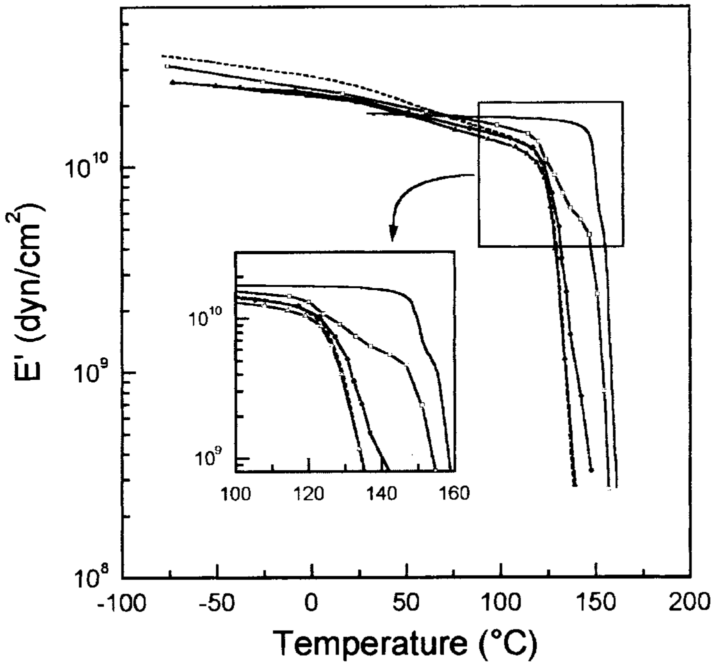


Figure 3. Temperature dependence of the storage modulus (E′) of PEN/PC blends: (- - -) PEN; (—) PC; (-Δ-) 85/15; (-●-) 75/25; (-□-) 50/50.

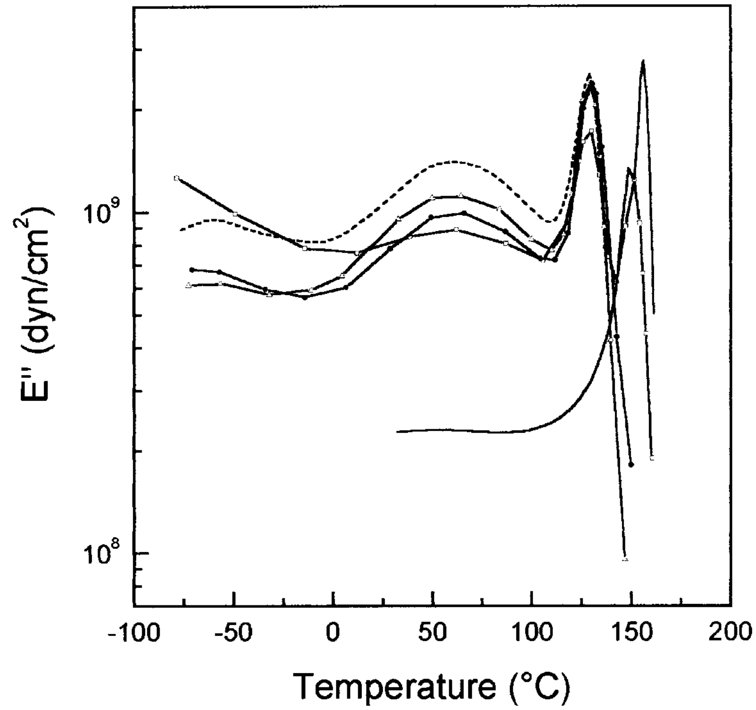


Figure 4. Temperature dependence of the loss modulus (E″) of PEN/PC blends: (- - -) PEN; (—) PC; (-Δ-) 85/15; (-●-) 75/25; (-□-) 50/50.

Thermal Properties

The glass transition temperatures were also determined by dsc and the results are presented in Table 3 and in Fig. 5. As shown, PEN/PC blends rich in PC exhibit two glass transitions indicating the presence of two amorphous phases. In contrast, only one T_g is apparent for blends containing 75% or more PEN, suggesting either a single mixed amorphous phase [16] or that the fine dispersion of PC in the PEN matrix (as shown in Fig. 7b) does not allow the obvious appearance of the PC phase. The results show also that the two glass transitions corresponding to those of the two phases (PC-rich and PEN-rich phase) are mutually displaced with the composition. The T_g of the PEN-rich phase does not shift so predominantly as the T_g of the PC-rich phase (insert, Fig. 5). These results are in agreement with the results of the extraction experiments where the presence of PEN in the soluble fraction is more obvious than the presence of PC in the insoluble.

The melting behavior of PEN in the PEN/PC blends is shown in Table 3. The melting temperature of PEN in the blends is shifted to lower temperatures with the increase of PC content. Although this T_m depression is not consistent with the composition, it may imply a possible miscibility of PEN

Table 3. Viscoelastic and Thermal Properties of Quenched and Annealed Samples of PEN/PC Blends

	DMA Results		DSC Results					
	T_g (°C)		T_g (°C)		$T_m{}^a$ (°C)	$T_m{}^b$ (°C) PEN Annealed at T_c		
PEN/PC Blends	PEN	PC	PEN	PC	PEN	214°C	220°C	230°C
100/0	128.6	–	122.7	–	266.5	256.8	260.4	267.4
90/10	n.t.	n.t.	122.8	–	260.7	249.1	255.3	264.0
85/15	129.0	–	123.4	–	256.1	248.4	253.9	262.9
75/25	130.0	–	123.8	–	253.8	249.4	254.1	262.3
50/50	128.7	151.0	124.1	143.1	260.5	250.0	254.5	–
25/75	n.t.	n.t.	124.5	144.9	259.2	251.7	253.9	–
15/85	n.t.	n.t.	126.5	146.3	256.8	256.4	256.1	–
100/0	–	156.0	–	148.6	–	–	–	–

[a] Obtained during the second heating scan.

[b] Obtained after annealing at the indicated temperatures T_c for 24 h.

n.t.: not tested.

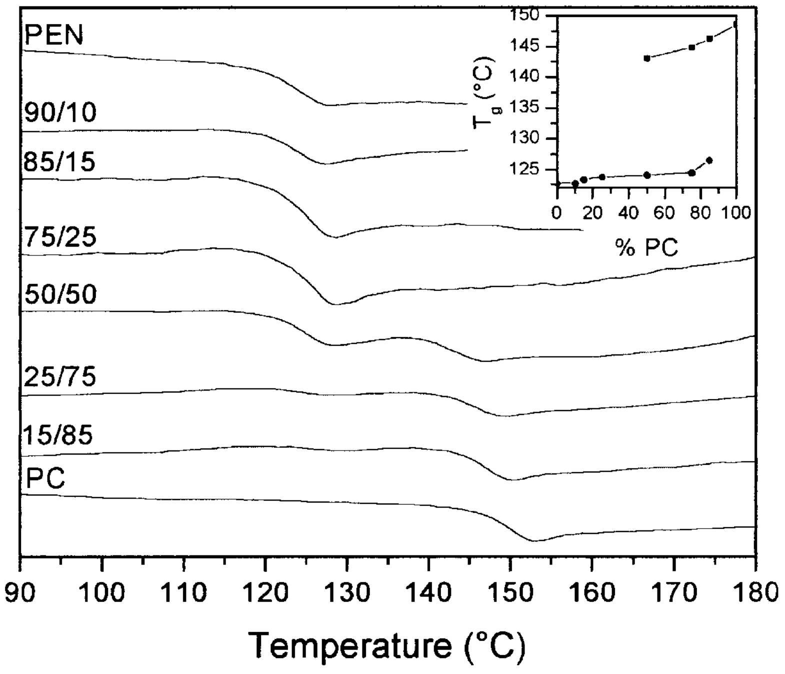


Figure 5. DSC thermograms of PEN/PC blends in the region of 90–180°C; insert: composition dependence of the PEN and PC T_g's in the blends.

with PC. In order to evaluate whether this depression is due to thermodynamic factors or is attributed to morphological parameters such as the crystal thickness, isothermal crystallization of the blends was performed at various temperatures, T_c's, and the subsequent T_m's were obtained. The results showed that the depression remains, but still no regularity with the composition is observed. In fact there is a considerable depression of the T_m of PEN in the blends that is not changing much with the increase of the PC content. So, no miscibility between PEN and PC was ascertained. The observed depression can be explained if we consider the formation of a PEN-PC copolymer during the reactive blending, which seems to be formed in a steady extent independently of the PC content. So, the observed T_m's of the blends could be assigned to a mixed phase of pure PEN with this copolymer independently of the composition, resulting in a steady T_m depression. The extent of PEN/PC copolymer formation due to transesterification occurring during melt mixing is examined by means of extraction experiments, microscopy, and spectroscopic methods.

Extraction Experiments

Solubility tests made for blend compositions PEN/PC 85/15, 75/25, and 50/50 testify to the presence of a copolymer formed during melt mixing under the processing conditions. The extraction experiments were performed in chloroform, at room temperature, in order to remove the unreacted PC from the blends. According to the results presented in Table 4, the amount of PC removed is less than the total PC amount in the blend. This indicates that a considerable amount of PC has reacted with PEN to form a copolymer that is no longer soluble in chloroform. Over the ranges studied, the removal of PC remains constant, revealing that the degree of transesterification achieved during blending is independent of the composition, which is in line with the DSC findings and the literature.[7,10]

The thermal behavior of the extracted blends was studied by DSC experiments. Comparison of the DSC trace (Fig. 6) of the blends, before

Table 4. Results of the Extraction Experiments on PEN/PC Blends in Chloroform at Room Temperature

PEN/PC Composition	Soluble Fraction (wt%) (Total Weight Regarded)	Soluble Fraction (wt%) (PC Weight Regarded)
100/0	0	0
85/15	9.6	64
75/25	16.4	65.6
50/50	31.5	63

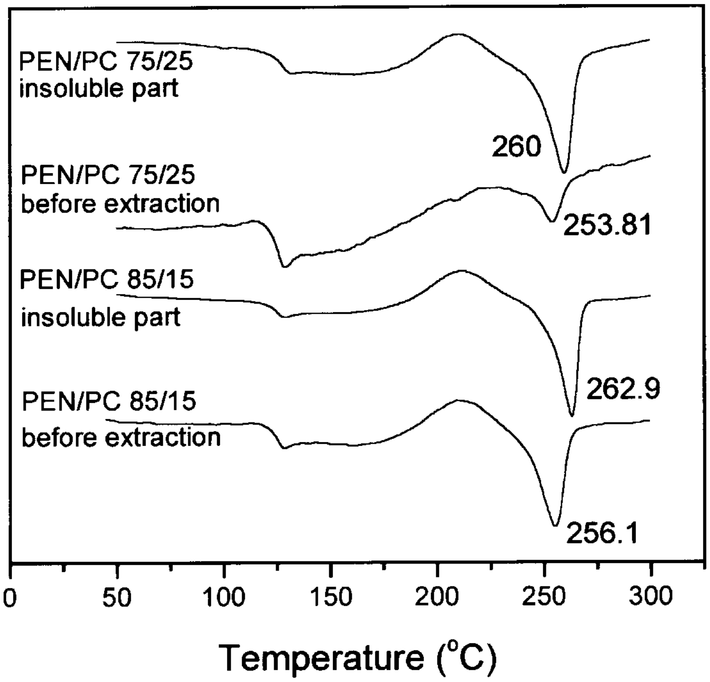


Figure 6. DSC thermograms of 85/15 and 75/25 PEN/PC blends in the region of 50–300°C before and after (insoluble parts) extraction with chloroform at room temperature (second heating scans).

and after the extraction of the soluble part, shows that the observed T_m depression is still evident after the removal of the PC-rich fraction. The above support the view that a PEN-PC copolymer coexists with the PEN phase, thus influencing its thermal behavior.

Morphology

Morphological examination of cryofractured surfaces of all blends is given in Fig. 7. All blends exhibit a biphasic character comprising through a homogenous dispersion of one phase into the other. The degree of dispersion becomes finer for the PEN- and PC-rich blends, as can be seen in the decreasing size of the particles consisting the minor phase. In 85/15 and 15/85 blends, the dispersed particles have an average diameter of less than 1 μm. Whether in 75/25 or 25/75 blends, the diameter increases slightly but still remains under 2 μm.

In the intermediate composition, it is not evident whether there is a clearly predominant phase, since phase inversion is likely to take place. The mixed ductile-glassy mode of fracture observed in all compositions examined justifies the high degree of adhesion implied by the tensile findings.

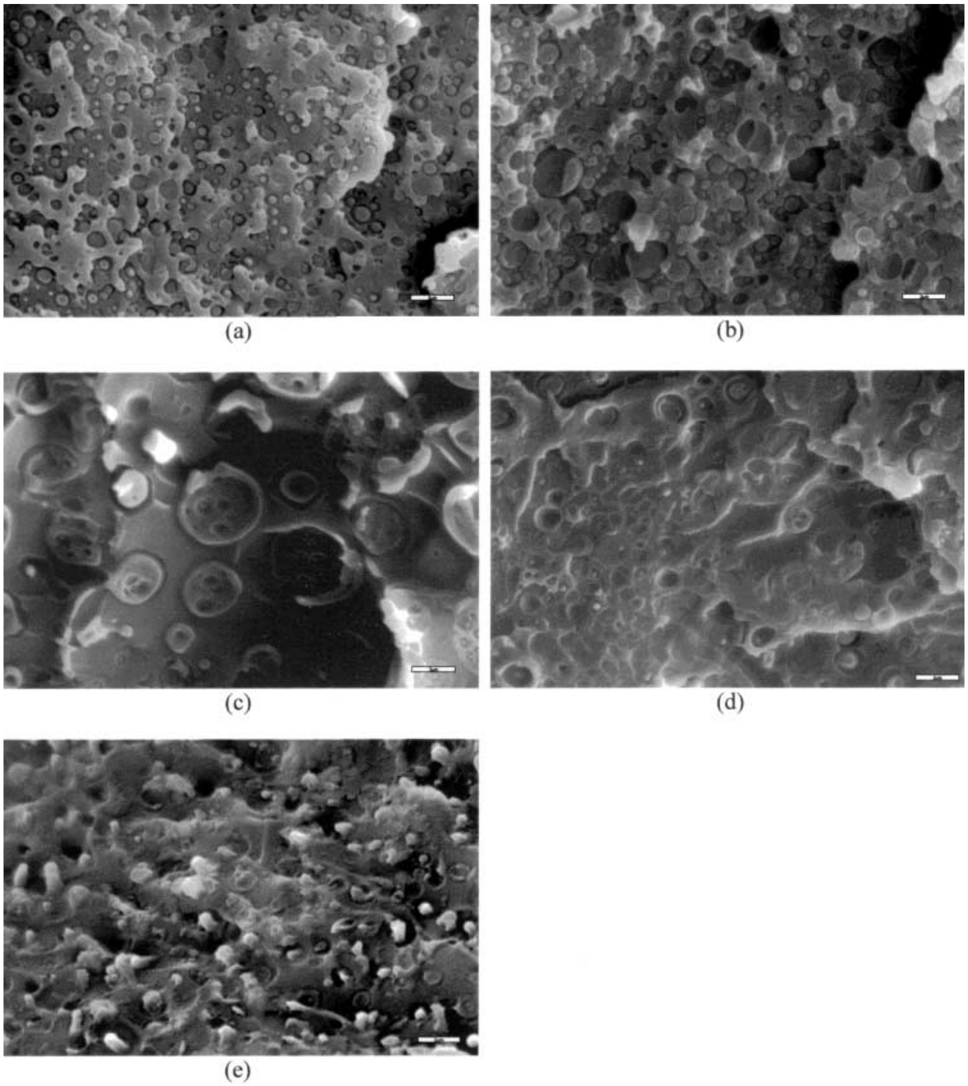
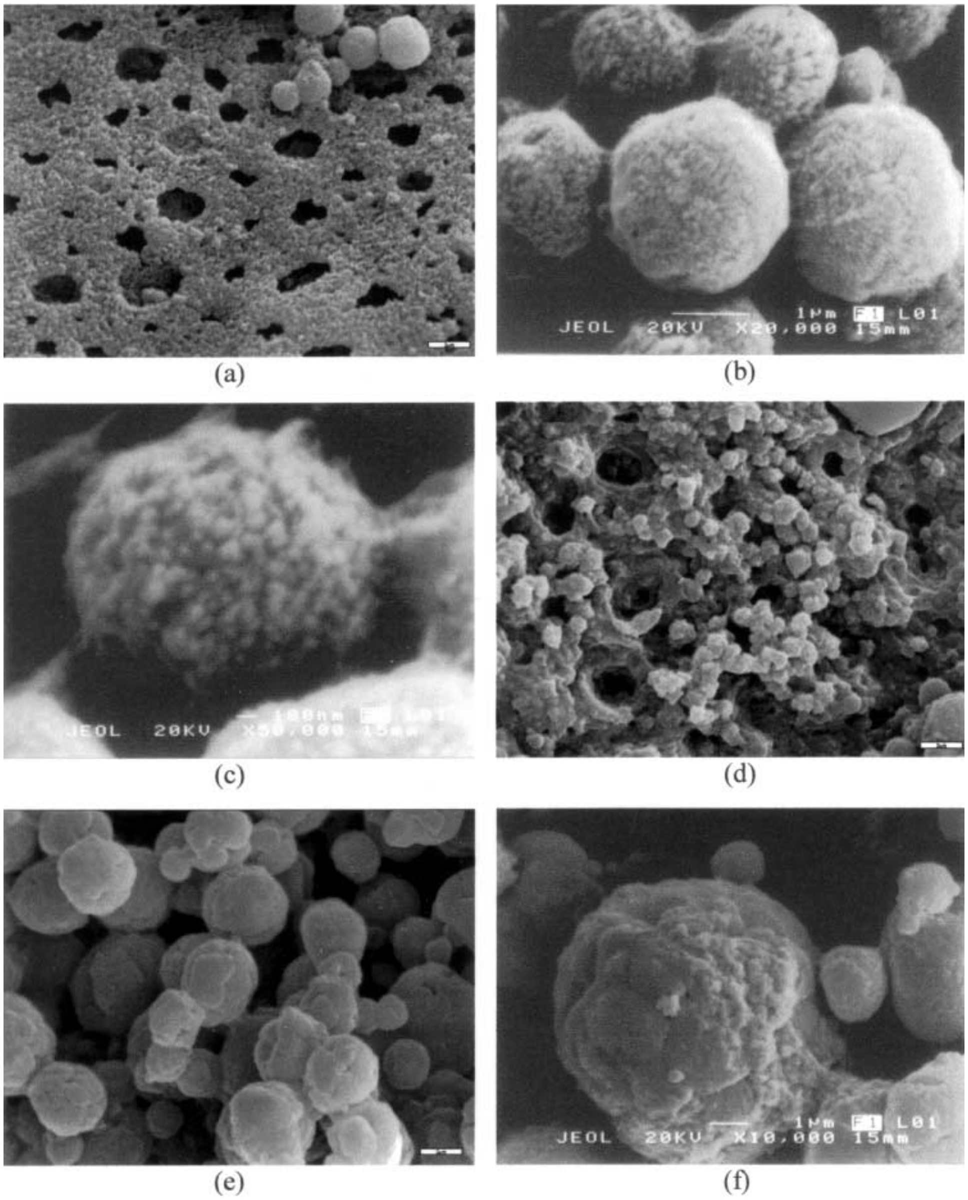

(a) (b) (c) (d) (e)

Figure 7. SEM micrographs of cryofractured PEN/PC blends: (a) 85/15; (b) 75/25; (c) 50/50; (d) 25/75; (e) 15/85. (Bars equal to 2 μm.)

Selective etching of PC from the cryofractured specimens by chloroform and subsequent SEM examination produced the micrographs of Fig. 8. As can be seen, the removal of PC leaves an abundant surface, with some particles left and dispersed holes of size analogous to that of the dispersed particles before etching. The 50/50 composition does not disintegrate, but it is evident that PEN is not clearly the matrix anymore.

In Figs. 8b and 8c, the texture of the remaining particles in the etched 85/15 sample is shown in detail. As seen there, the sponge-like particles must



(a) (b) (c) (d) (e) (f)

Figure 8. SEM micrographs of cryofractured and etched with chloroform at room temperature PEN/PC blends: (a) 85/15 (× 5000); (b) 85/15 (× 20,000); (c) 85/15 (× 50,000); (d) 75/25 (× 5000); (e) 50/50 (× 5000); (f) 50/50 (× 10,000).

be PC grafted on the PEN matrix, which could not be removed by the extraction. This is something that was not observed to the same extent for the remaining particles in the etched 50/50 sample (Fig. 8f), where the particles have different appearance.

FT-IR AND FT-RAMAN EXAMINATION

The presence of PC in the insoluble fraction after the extraction can be qualitatively estimated by spectroscopic methods. In the indicative FTIR spectra of the 75/25 blend after the extraction (Fig. 9), a peak at 1776 cm^{-1}, characteristic of the carbonyl group in PC, is observed. Additionally, in the same figure the spectra of the extracted part, i.e., the soluble part of the blend, shows that the removal of PC has washed away PEN segments as well. This is shown by the characteristic absorption at 1725 cm^{-1} of the carbonyl group of PEN. These observations indicate the formation of a PEN-PC copolymer during melt mixing, which according to the character of the constituent that dominates, is soluble or not in chloroform.

The FT-Raman spectrum of the soluble fraction of the 85/15 PEN/PC blend after its extraction with chloroform at room temperature further justifies the suggestion of copolymer formation (Fig. 10). The spectrum of the soluble fraction clearly contains the peak at 1721 cm^{-1} due to the carbonyl group of PEN and the peaks at 1390 and 1634 cm^{-1} due to the naphthalate units.

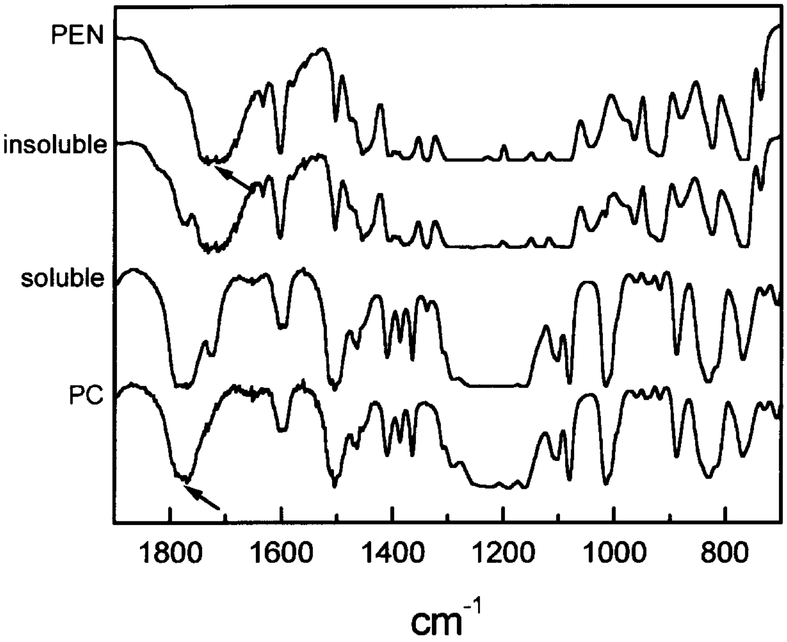


Figure 9. FTIR spectra of the soluble and insoluble fractions of the 75/25 PEN/PC blend after the extraction with chloroform at room temperature and of the pure constituents.

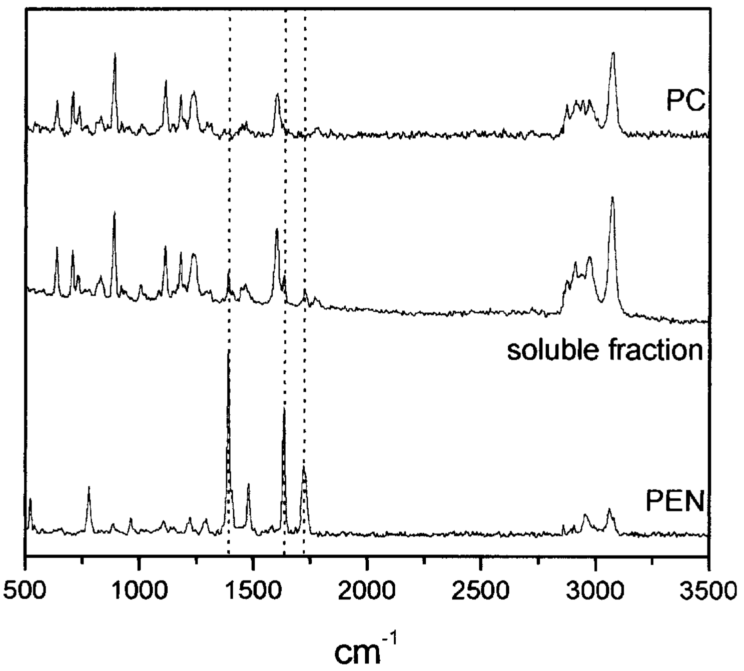


Figure 10. FT-Raman spectra of the soluble fraction of the 85/15 PEN/PC blend after the extraction with chloroform at room temperature and of the pure constituents.

NMR EXAMINATION

^{1}H NMR analysis of the soluble and insoluble fractions can give evidence of the formation of PEN/PC block copolymers. In fact, the presence of PEN in the soluble fraction and of PC in the insoluble one proves the formation of block copolymers. If no reactions had occurred between PC and PEN, they would have been found completely in the soluble and insoluble fractions, respectively.[21] The NMR spectrum of 85/15 PEN/PC blend prepared at 290°C and 10 min melt mixing is presented in Fig. 11a. The insoluble fraction of this blend after the extraction gave the spectrum shown in Fig. 11b, where we can observe the signals of the aromatic protons of bisphenol A (7.1–7.4 ppm) and the methyl protons of bisphenol A (1.7 ppm). These can be considered as an evidence for the grafting of PC on the PEN-matrix.

CONCLUSION

Melt mixing of PEN with PC resulted in blends with improved mechanical properties related to the pure constituents. Blends at various compositions were studied to understand their compatibility behavior. DMA and morphological studies performed revealed the biphasic character of these

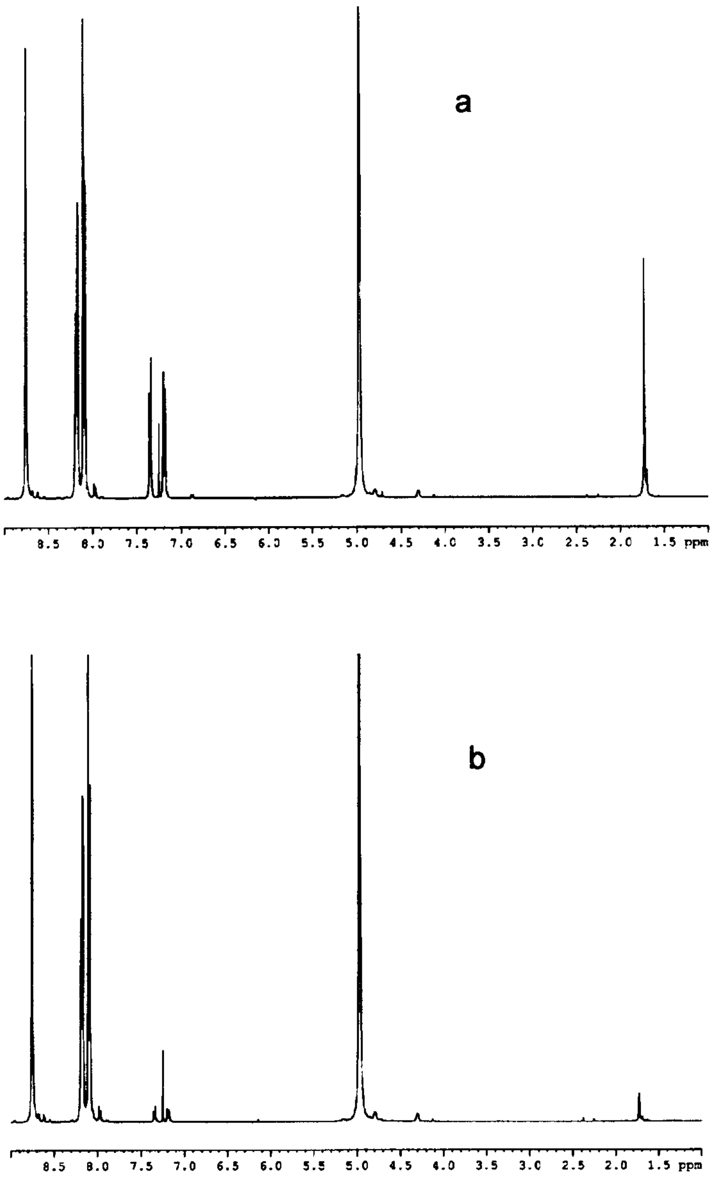


Figure 11. NMR spectra of the 85/15 PEN/PC blend (a) before the extraction with chloroform at room temperature; (b) insoluble fraction.

blends. Tensile testing shows improved mechanical properties for the PEN-rich blends, denoting thus the very good adhesion between two phases. Extraction experiments and subsequent characterization using spectroscopic techniques showed the existence of both polymers in the soluble and insoluble fractions, supporting the view that during the melt processing transesterification reactions occurred resulting in a PEN/PC copolymer formation. Although its minor amount, the copolymer seems to act as effective compatibilizer for the initially immiscible PEN/PC blends, probably because it formed and stayed at the interface.

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