Polyester Resin as a Compatibilizing Agent for Some Polymeric Blends

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ABSTRACT: Dielectric and viscosity techniques were used to determine the degree of the compatibility of poly(methyl methacrylate)/polycarbonate, poly(methyl methacrylate)/polystyrene, and polycarbonate/polystyrene blends in different ratios (25/75, 50/50, and 75/25 w/w). The effect of the addition of 5, 10, and 20% concentrations of the prepared polyester resin [poly(butylene terephthalate adipate)] on the compatibility of these blends was studied. The dielectric properties were measured over a frequency range (from 100 Hz to 100 kHz) at various temperatures covering the glass-transition temperatures of the polymers used (from 30 to 170°C). It was found from the dielectric and viscosity measurements that the addition of 10% polyester to poly(methyl methacrylate)/polycarbonate, 20% polyester to poly(methyl methacrylate)/polystyrene, and 5% polyester to polycarbonate/polystyrene blends enhanced the degree of compatibility of such blends. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 108: 833–844, 2008

Key words: blends; compatibility; dielectric properties; polyesters; viscosity

INTRODUCTION

Amorphous polymers such as poly(methyl methacrylate) (PMMA), polystyrene (PS), and polycarbonate (PC) are attractive for many engineering applications because of their excellent transparency, high moduli, and relative ease of processing, but they exhibit shortcomings in some properties.

Polymer blending is an efficient way to improve the properties of the individual polymers. Voluminous work[1,2] has been carried out in this area to understand the mechanical, thermal, electrical, and processing behavior of these blends, which can be correlated to changes in the structure and phase morphology of the blends. The major problem is the lack of miscibility, which leads to the dispersion of large minor phase domains in the matrix of the blends. The multicomponent polymer systems have been found to be incompatible for various reasons, such as the absence of any specific interaction between the blend constituents, dissimilarity in their structures, and broad differences in their viscosities, surface energy, or activation energy of flow and polarity.

Effective compatibilization is needed to improve the interfacial adhesion between the phase components. However, the phenomenon of compatibility can be induced in an immiscible polymer–polymer pair in a binary system by the introduction of a third component that will either interact with both phases chemically or have a specific interaction with the other.[3–6] It may be emphasized here that the function of the third component is to reduce the interfacial tension between the two phases, increase the surface area of the dispersed phase, promote adhesion between the phase components, and stabilize the dispersed phase morphology. Therefore, it is necessary to introduce a compatibilizing agent that can act as an interface between the constituents of the polymer blend. The effect of polyester resins as compatibilizing agents on the degree of compatibility of many types of polymers has been studied.[7,8]

A variety of experimental methods have been used to study the blend compatibility. These methods include viscometric measurements,[9] dynamic mechanical analysis, electron microscopy, differential scanning calorimetry (DSC),[10–12] infrared (IR) spectroscopy, dielectric permittivity,[13,14] and ultrasonic measurements.[15,16]

The aim of this investigation was to prepare a suitable polyester that could act as a compatibilizing agent for series of PMMA/PC, PMMA/PS, and PC/PS blends. The degree of compatibility was studied through dielectric and viscosity techniques.

EXPERIMENTAL

Materials

Reagent-grade dimethyl terephthalate, 1,4-butanediol, adipic acid, chloroform, and zinc acetate as the

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transesterification catalyst were obtained from Merck (Darmstadt, Germany) and used as received.

PS was supplied by BASF (Ludwigshafen, Germany) under the technical name 168N-003 Glass Klar (weight-average molecular weight = 25 × 10^4 g/mol).

PC of bisphenol A (Makrolon 2808) was provided by Bayer (Leverkusen, Germany; weight-average molecular weight = 28 × 10^3 g/mol).

PMMA was obtained from Röhm (Darmstadt, Germany) in a granular form with a weight-average molecular weight of 15 × 10^4 g/mol.

Methods

Synthesis of the saturated polyester resin

Poly(butylene terephthalate adipate) was prepared by a two-stage melt polycondensation method (esterification and polycondensation). A three-necked flask equipped with a condenser, nitrogen inlet tube, and magnetic stirrer was charged with 194 g (1 mol) of dimethyl terephthalate and 198 g (2.2 mol) of 1,4-butanediol. The reaction mixture was heated to 150°C for 3 h. This first step (esterification) was considered to be completed after the collection of a theoretical amount of methanol, which was removed from the reaction mixture by distillation and collected in a graduate cylinder. In the second step of polycondensation, 146 g (1 mol) of adipic acid was added, the mixture was stirred for another 5 h, and the temperature was increased from 160 to 200°C at a rate of 10°C/h. The reaction was followed by the determination of the acid value of the polyester until it reached 42 mg of KOH/g.

The carboxyl end group content (—COOH) of the resin was determined as follows. About 0.2 g of polyester was dissolved in chloroform at room temperature, and the solution was titrated with standard KOH in methanol (N/10) and phenolphthalein as an indicator.

Preparation of the polymer blend by the solution-casting technique

The required solution was prepared by 1 g each of PMMA, PC, and PS being dissolved separately in 100 mL of chloroform. Films of polymer blends (PMMA/PC, PMMA/PS, and PC/PS) were prepared by the casting of mixed solutions of different ratios (0/100, 25/75, 50/50, 75/25, and 100/0) with and without 5, 10, or 20 wt % of the prepared polyester on a glass plate with slow drying at room temperature. Care was taken to control the uniform thickness of the films for all compositions. Clear films were obtained with thicknesses ranging from 0.10 to 0.15 mm.

Techniques

IR spectra were recorded on a Jasco (Tokyo, Japan) FT/IR 300E Fourier transform infrared spectrometer.

The proton nuclear magnetic resonance (^{1}H-NMR) spectrum was run at 260 MCPS on a JEOL (Tokyo, Japan) Ex-270 NMR spectrometer.

The molecular weight determination was done by gel permeation chromatography (GPC). GPC was carried out with an Agilent Technologies (Walden Bornn, Germany) 1100 series equipped with two Styragel columns (10^2 and 10^4 Å) and an Agilent G 1362 refractive index detector. Tetrahydrofuran was used as the eluent at a flow rate of 1 mL/min. The columns were calibrated by means of PS (an internal standard was used).

DSC was performed on a PerkinElmer (Waltham, MA) DSC-7 at a scanning rate of 10°C/min under a nitrogen atmosphere.

The viscosities of the polymer blend solutions in chloroform were measured with the AV 350 from Schott Gerate (Hofheim, Germany). Stock solutions of each system were prepared and diluted to yield five concentrations (1, 0.8, 0.6, 0.4, and 0.2 g/100 mL) made by the addition of appropriate amounts of chloroform to the stock solutions. In this case, the liquid meniscus was sensed optoelectrically at the measuring levels. These precision devices always ensured the high accuracy of the capillary viscometer. The viscosity-measuring unit (Schott Gerate) was connected to a CK100 flow-through cooler to control the temperature to an uncertainty of ±0.1°C, and the measurements were carried out at 30°C.

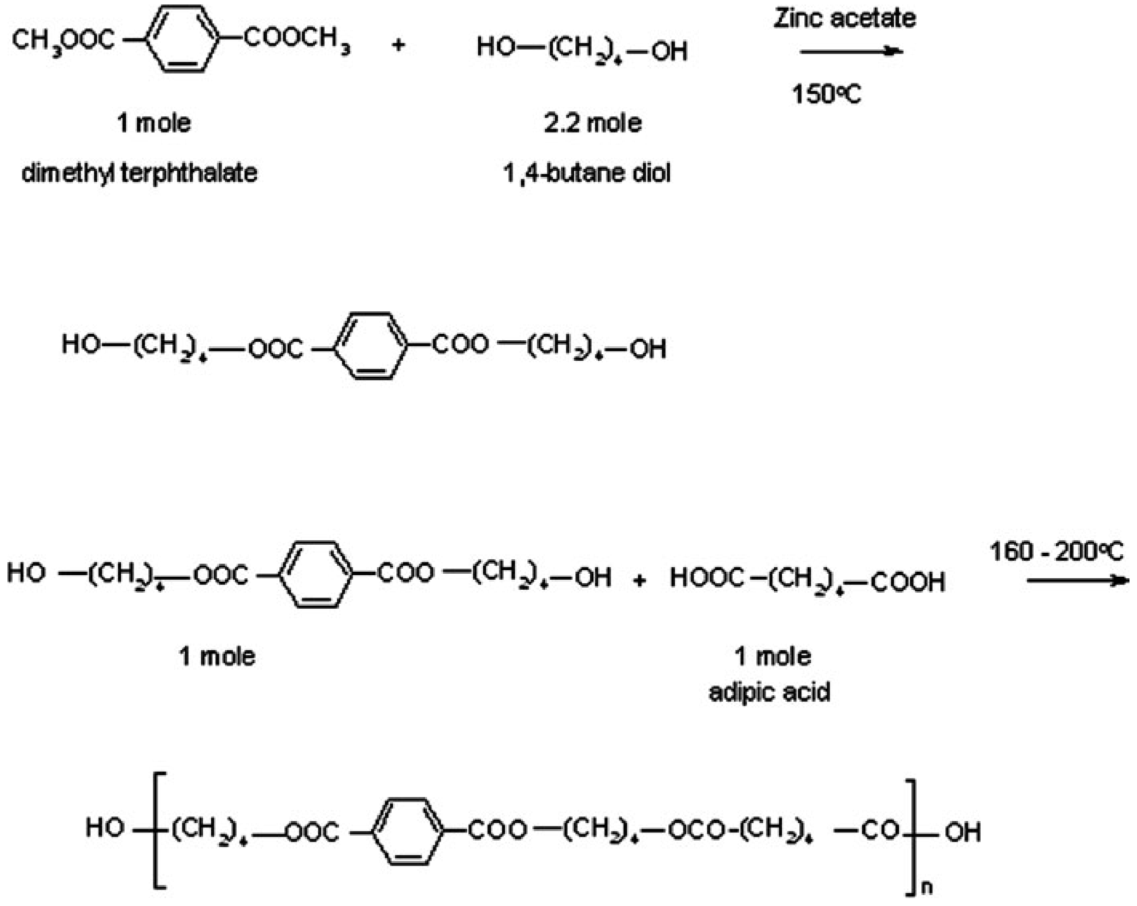
Dielectric measurements were carried out in the frequency range of 100 Hz to 100 kHz with an AG-411 B inductance capacitance resistance (LCR) meter (Ando Electric, Ltd., Tokyo, Japan). The capacitance, loss tangent (tan δ), and alternating-current resistance were measured directly from the bridge, from which the permittivity (ε'), dielectric loss (ε''), and direct-current resistance were determined. The cell used was a microcapacitor consisting of two glass plates (thickness = 0.050 mm and area = 1 cm^2). The experimental error in ε' and ε'' was found to be ±3 and ±5%, respectively. The temperature was controlled by the placement of the cell in a digital oven, and the experimental error in the temperature was ±0.1°C.

RESULTS AND DISCUSSION

The polyester was prepared according to Scheme 1.

The prepared polyester was a light yellow, hard resin with a number-average molecular weight of 1327 g/mol and a glass-transition temperature (T_g) of −20°C.

The chemical structure of the prepared polyester was characterized with IR and ^{1}H-NMR spectroscopy. The IR spectrum of the prepared polyester (Fig. 1)



Scheme 1 Esterification as the first step (top) and polycondensation as the second step (bottom).

indicates the presence of an intense, strong band at 1720 cm^{-1} characteristic of the stretching frequency of acid and ester carbonyl groups. The broad band appearing at 3440 cm^{-1} stands for OH groups, and the band at 2960 cm^{-1} is due to the stretching frequency of CH_2. The strong, broad C—O—C stretching vibrations appear from 1275 to 1100 cm^{-1}, whereas the bands at 730, 880, and 935 cm^{-1} are indicative of the bending frequency of the aromatic —CH group.

The ^{1}H-NMR spectrum (Fig. 2) shows a signal at δ = 8.01 ppm indicating the presence of the four aromatic protons of the terephthalic residue. The two triplets at δ = 4.4–3.5 ppm are characteristic of the methylene protons of $O{=}CO{-}CH_2$ and $CH_2{-}OH$ of 1,4-butanediol. The peaks at δ = 2.21 ppm are due to methylene protons adjacent to the carbonyl group of adipic residue $OCCH_2 \cdots CH_2CO$. The central (internal) methylene groups of butane diol overlapping with the internal methylene groups of adipic acid ($-CH_2-CH_2-$) appear at δ values of 1.45 and 1.40 ppm.

Compatibility study

To evaluate the compatibility of the PMMA blends with PC and PS (system I) and the PC/PS blends (system II), the dielectric and viscosity measurements were studied before and after the addition of 5, 10, or 20% polyester resin. From the dielectric measurements, T_g and ε' were determined. The obtained data [ε' at 100 Hz, T_g, and the intrinsic viscosity (η_i)] are listed in Tables I–III.

I: PMMA blended with PC and PS

ε' and ε'' were measured in the frequency range of 100 Hz to 100 kHz and at temperatures ranging

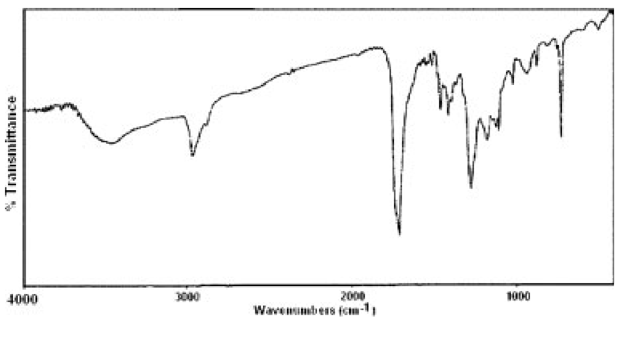


Figure 1 IR spectrum for the prepared polyester resin.

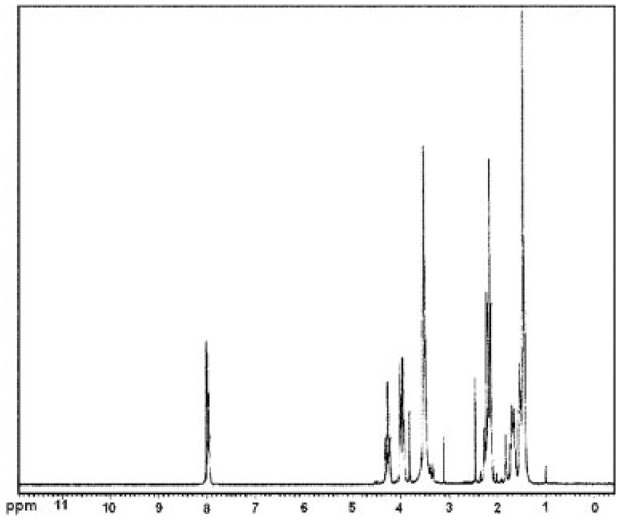


Figure 2 ^{1}H-NMR spectrum for the prepared polyester resin.

from 30 to 170°C for PMMA/PC and PMMA/PS blends with different ratios (0/100, 25/75, 50/50, 75/25, and 100/0) before and after the addition of the polyester with three different ratios (5, 10, and 20%) as a trial to improve the compatibility of such blends.

The best results were obtained by the addition of 10 and 20% polyester to PMMA/PC and PMMA/PS blends, respectively. For such concentrations, ε' and ε'' for PMMA/PC and PMMA/PS before and after the addition of polyester at 30 and 170°C are plotted graphically versus the frequency, as shown in Figures 3–6. From these figures, it is clear that ε' decreases with the percentage of PMMA increasing, whereas it decreases with the applied frequency increasing, which shows an anomalous dispersion.

From these figures, it is also clear that the curves relating ε'' and the logarithm of the frequency are broader than a Debye curve,[17] indicating that more than one relaxation process is present. These processes could be attributed to mechanisms related to the main chain and its related motions.[18,19] One of the expected mechanisms is related to the Maxwell–Wagner effect, which usually occurs at a lower frequency range for heterogeneous systems. The origin of such a process is an alternating current that is in phase with the applied potential because of the difference in the permittivities and resistivities of the constituents in the blend. Another mechanism is expected to be at a higher frequency range and can be ascribed to local molecular motions rather than the main-chain motion for temperatures below T_g, as the main-chain motion is frozen before T_g. Above

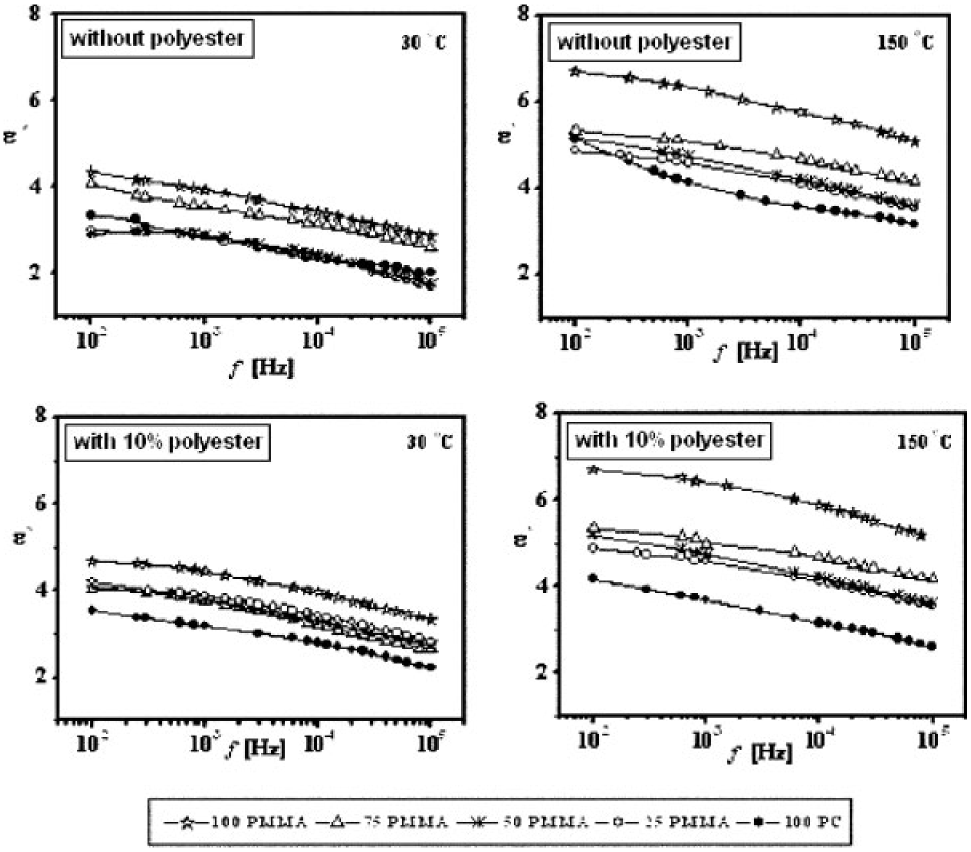


Figure 3 ε' versus the frequency (*f*) for the PMMA/PC blends at 30 and 150°C with and without polyester.

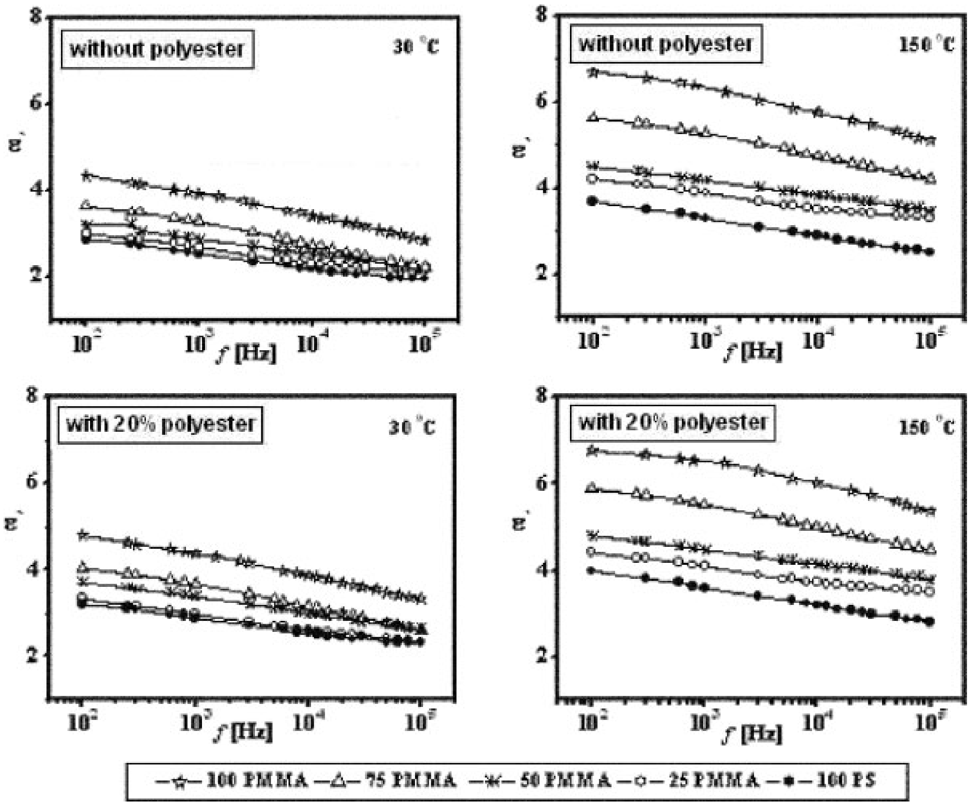


Figure 4 ε′ versus the frequency (*f*) for the PMMA/PS blends at 30 and 150°C with and without polyester.

the glass transition, this process can be attributed to the combination of the large-scale mobilization of the chains, that is, the glass–rubber relaxation process, and a contribution of the motion of the large aggregates caused by the movement of the main chain.

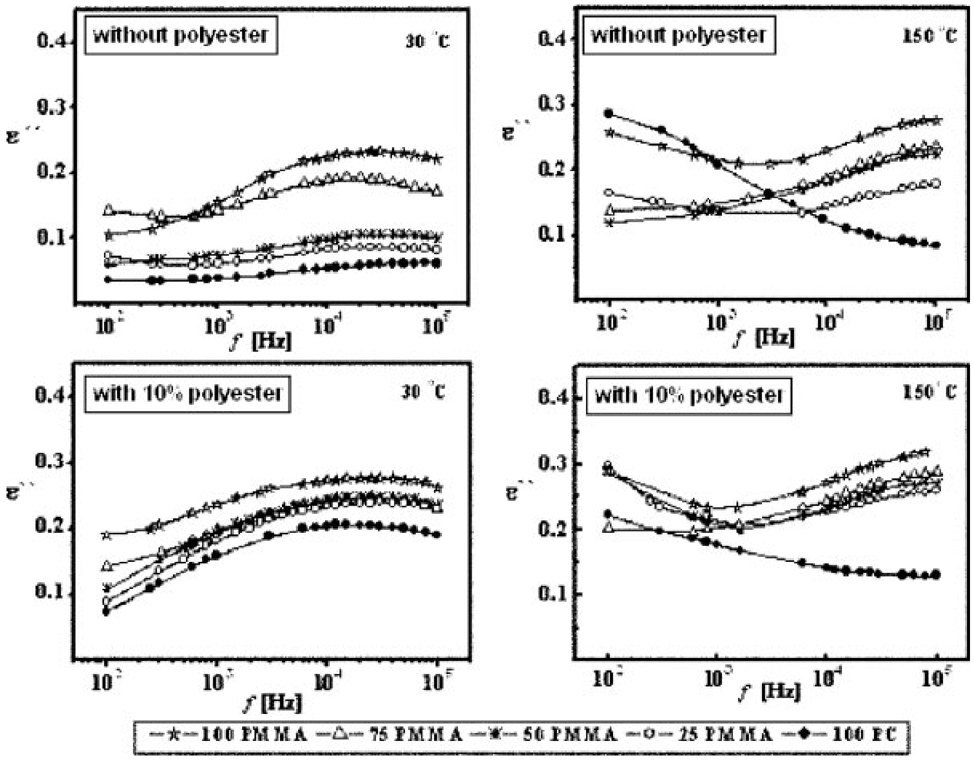


Figure 5 ε″ versus the frequency (*f*) for the PMMA/PC blends at 30 and 150°C with and without polyester.

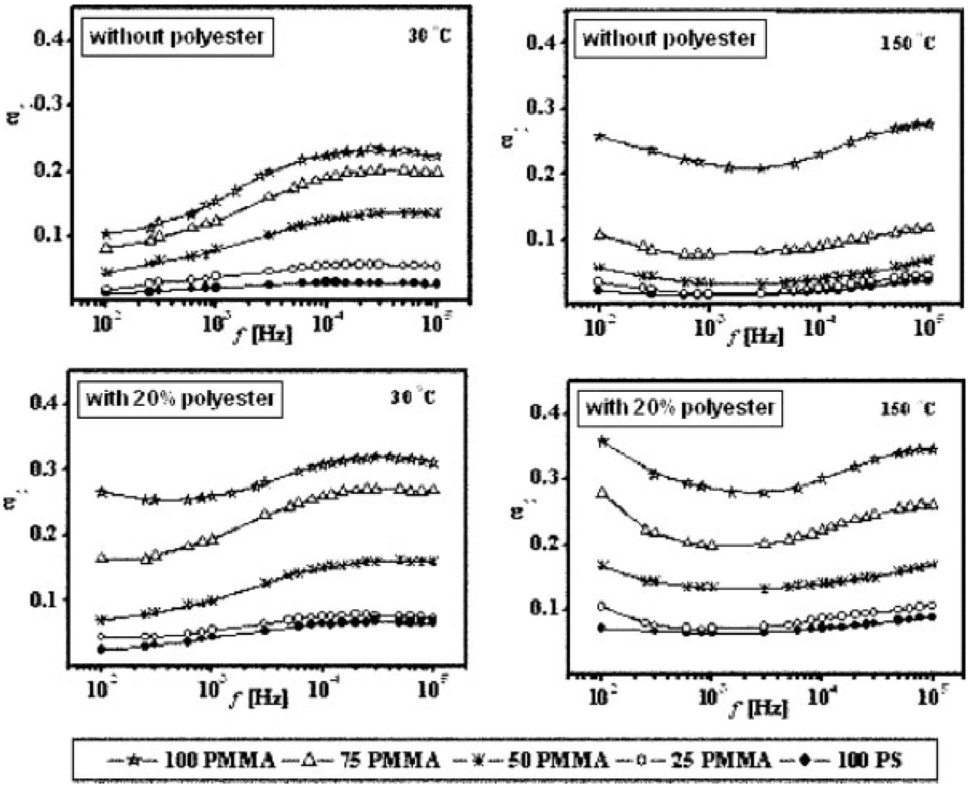


Figure 6 ε″ versus the frequency (*f*) for the PMMA/PS blends at 30 and 150°C with and without polyester.

It is notable that the addition of 10 and 20% polyester increases both ε′ and ε″ as polyester is a polar compound that can add some polar dipoles to the blends, leading to the increase in both ε′ and ε″.

Dielectric measurements

T_g. The variation of tan δ versus the temperature (the isochrone temperature scans) is illustrated graphically at 100 Hz in Figure 7. From this figure, it is seen that tan δ shows a broad peak. This result is considered to be good evidence for the presence of superimposed processes due to the orientation of the main chain and its related motions, which may be attributed to the large-scale mobilization of the chains, that is, the glass–rubber relaxation. In addition to the glass–rubber relaxation process, a contribution of the side-chain motion may be present. The values of tan δ at the peaks of these curves, which represent T_g, were obtained and are listed in Tables I and II.

The relation between the values of T_g and the PMMA content before and after the addition of 10% polyester in the case of PMMA/PC blends and 20% polyester in the case of PMMA/PS blends is given in Figures 8 and 9, respectively. From these figures, it is clear that the values of T_g for blends free from polyester do not coincide on the line connecting the T_g values of the two individual polymers. This indicates that the two polymers under investigation are incompatible. On the other hand, for PMMA/PC blends containing 10% polyester, this relationship is represented by a straight line. This linearity indicates that the addition of 10% polyester improves to some extent the compatibility of such blends. From Table I, one can conclude that the addition of 20% polyester can lead to the same conclusion: an improvement in the compatibility of PMMA/PC. Therefore, it can be concluded that the addition of 10% polyester is convenient for solving the problem of phase separation of such blends. The values of PMMA/PS blends before and after the addition of polyester are listed in Table II. Figure 9 represents the variation of T_g versus the PMMA content for PMMA/PS blends free from polyester and those containing 20% polyester. From this figure, it can be seen that the linear relationship between T_g and the PMMA content in the blends occurred at a concentration equal to 20% polyester.

ε′. The relationship between the values of ε′ at a frequency of 100 Hz and the content of PMMA before and after the addition of 10% polyester for PMMA/PC blends and 20% polyester for PMMA/PS blends is represented in Figures 8 and 9. From these figures, it is clear that the values of ε′ deviate from the line connecting the values of ε′ for the two individuals in the case of the blends free from polyester. This devi-

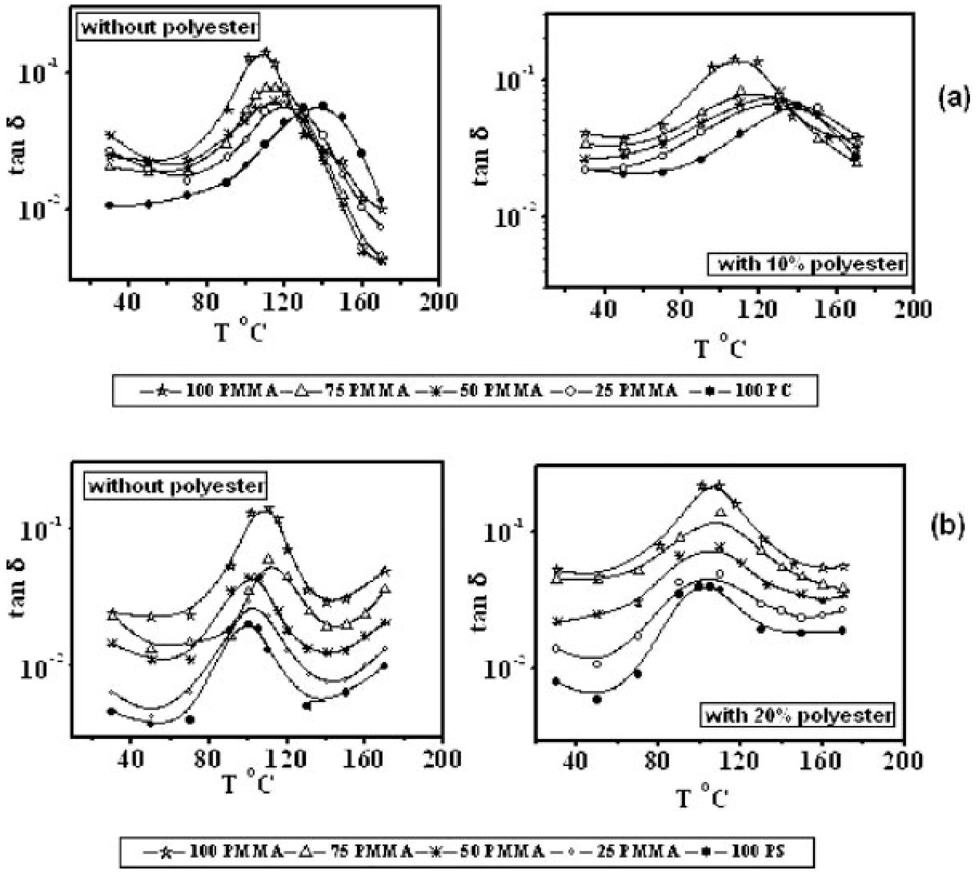


Figure 7 Variation of tan δ versus the temperature (*T*) for (a) the PMMA/PC blends and (b) the PMMA/PS blends with and without polyester.

TABLE I
ε', T_g, and η_i Values of PMMA/PC Blends with Different Polyester Contents

PMMA content	ε' at 100 Hz	T_g (°C)	η_i
0% polyester			
100	4.35	106.5	1.01
75	3.90	112.8	0.65
50	3.50	114.8	0.49
25	3.40	121.0	0.40
0	3.33	140.0	0.48
5% polyester			
100	4.65	106.0	1.22
75	3.66	114.0	0.98
50	3.61	122.0	0.71
25	3.37	126.0	0.44
0	3.40	140.5	0.49
10% polyester			
100	4.67	106.0	1.47
75	4.21	116.0	1.23
50	4.12	126.0	1.00
25	4.05	132.0	0.78
0	3.54	140.0	0.53
20% polyester			
100	4.78	105.8	1.49
75	4.50	111.9	1.64
50	4.25	134.7	1.27
25	4.10	139.0	0.97
0	3.70	140.0	0.60

TABLE II
ε', T_g, and η_i Values of PMMA/PS Blends with Different Polyester Contents

PMMA content	ε' at 100 Hz	T_g (°C)	η_i
0% polyester			
100	4.35	106.5	1.01
75	3.53	103	1.05
50	3.20	100	1.15
25	3.65	99	1.22
0	2.84	98	1.73
5% polyester			
100	4.65	106.6	1.22
75	3.66	104.9	1.1
50	3.61	102.0	1.22
25	3.37	101.0	1.56
0	2.96	99.5	1.92
10% polyester			
100	4.67	106.1	1.47
75	4.25	105.0	1.60
50	3.70	103.0	1.83
25	3.25	101.0	1.99
0	2.98	100.0	2.17
20% polyester			
100	4.80	107	1.49
75	4.41	106	1.71
50	4.01	104	1.88
25	3.70	102	2.20
0	3.23	100	2.38

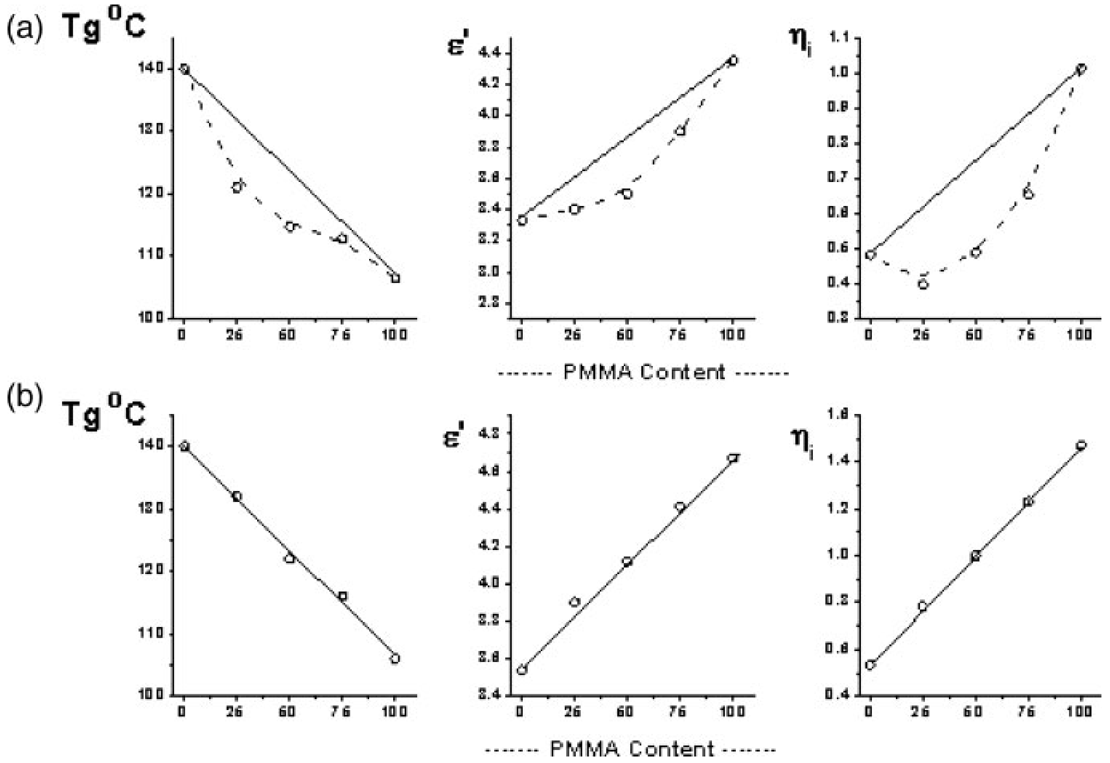


Figure 8 Variation of T_g, ε', and η_i versus the PMMA content in the PMMA/PC blends (a) without and (b) with 10% polyester.

ation, which is found to range from 12 to 20%, is considered to be higher than the experimental error in the ε' values (2–3%). This deviation is reduced to 0.7–3% by the addition of 10% polyester to the PMMA/PC blend and 20% polyester to the PMMA/PS blend, which is considered to be within the

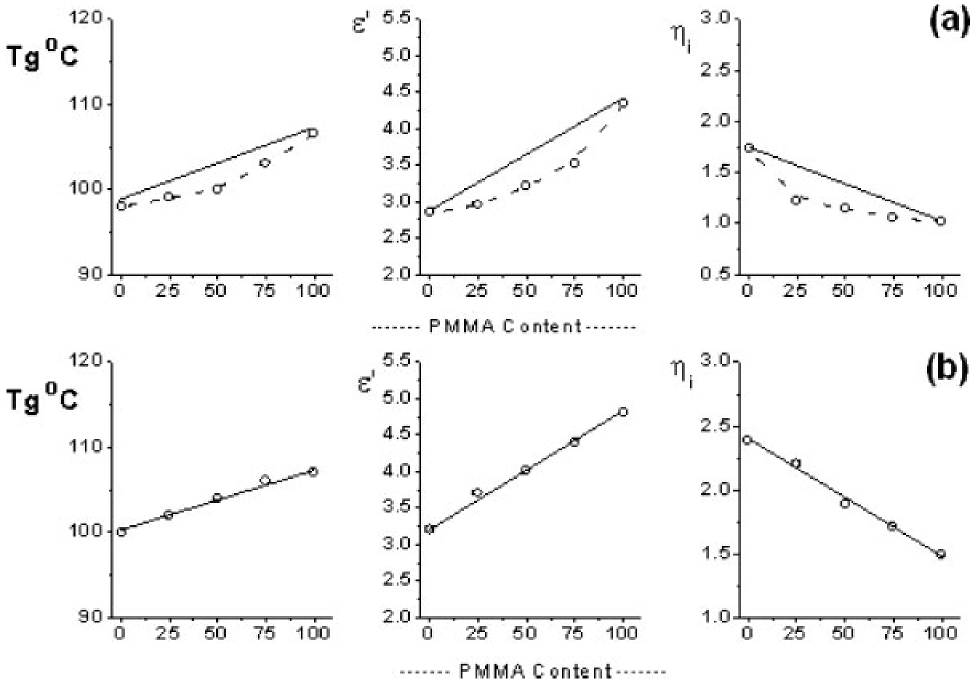


Figure 9 Variation of T_g, ε', and η_i versus the PMMA content in the PMMA/PS blends (a) without and (b) with 20% polyester.

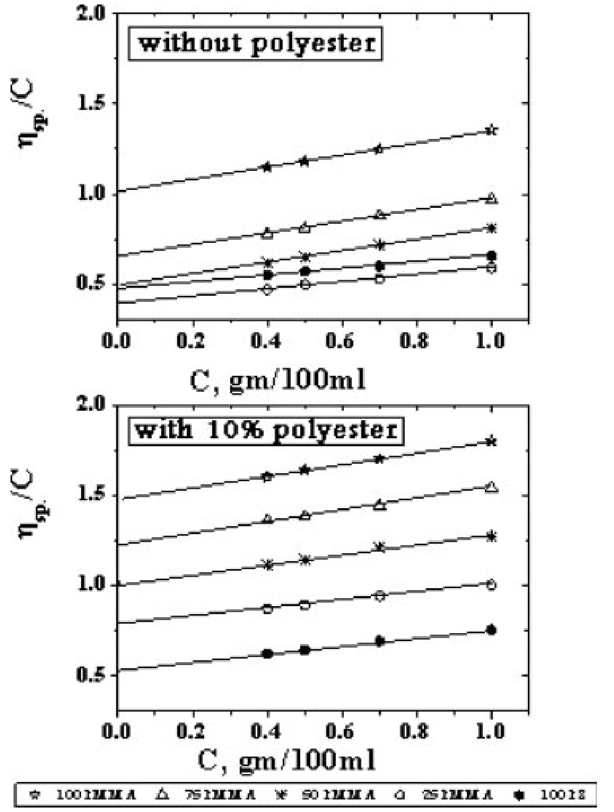


Figure 10 Relation between η_{sp} and C (g/100 mL) of the PMMA/PC blends with and without polyester.

experimental error. This result could indicate that the addition of polyester in such concentrations improves to some extend the degree of compatibility of such blends.[13]

Viscosity measurements

A viscosity technique has been applied to investigate the miscibility of the aforementioned blends in dilute solutions. The nature of the viscosity of dilute polymer solutions is based on analyzing hydrodynamic properties related to the movement of macromolecules in solutions. The two-phase structures of polymer mixtures and deformation of drops in flow mixtures depend not only on the component ratio but also on the value of the viscosity, which is estimated from the degree of deviation of η_i from the line connecting the two individuals. The degree of compatibility of the polymer blends can be deduced from the following equation:[20]

$$(\eta_{sp}/C) = \eta_i + K\eta_i^2 C$$

where η_{sp} is the specific viscosity, C is the concentration (g/100 mL), and K is a constant. A linear relationship is obtained by the plotting of η_{sp}/C against concentration C. From the intercepts of these lines

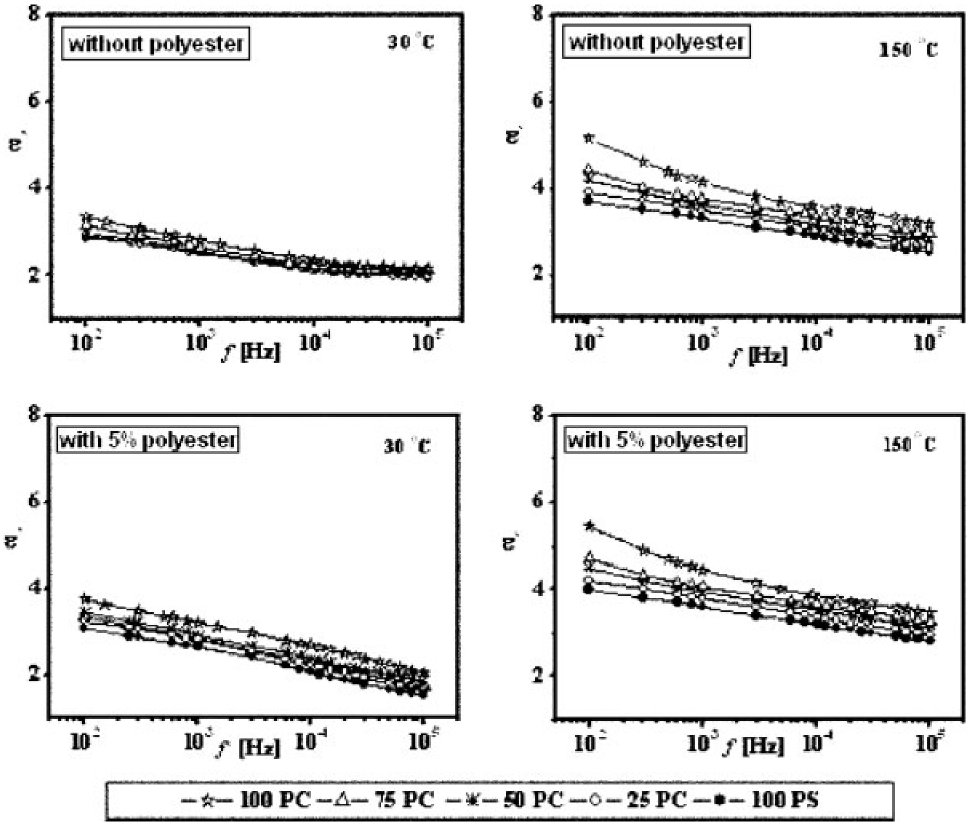


Figure 11 ε′ versus the frequency (*f*) for the PC/PS blends at 30 and 150°C with and without polyester.

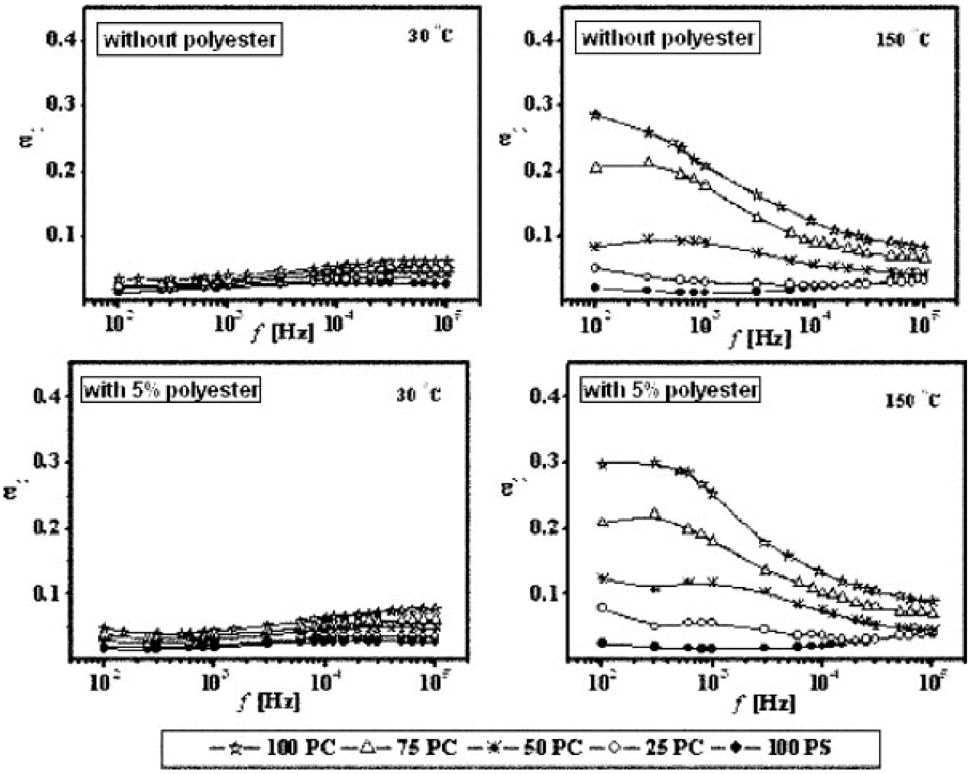


Figure 12 ε″ versus the frequency (*f*) for the PC/PS blends at 30 and 150°C with and without polyester.

with the *y* axis, η_i can be obtained.[21] An example of these measurements is illustrated graphically in Figure 10. The obtained values for η_i are listed in Tables I and II, and examples of the obtained data are illustrated in Figures 8 and 9 for PMMA/PC and PMMA/PS blends free from polyester and those containing 10 and 20% polyester, respectively. It is clear that the values of η_i do not coincide with the line connecting the two individuals for blends free from polyester. This deviation from linearity leads to some sort of incompatibility in such blends.[22] On the other hand, Figures 8 and 9 show that the values of η_i nearly coincide with the line connecting the two individuals for PMMA/PC blends containing 10 and 20% polyester respectively. The deviation from linearity does not exceed 2%, which lies within the experimental error. This means that these blends are compatible confirms the result obtained by ε′ and T_g mentioned previously.

II: PC/PS blends

ε′ and ε″ were measured in the same way as discussed previously before and after the addition of polyester in different concentrations. Examples of the obtained data are illustrated graphically in Figures 11 and 12. From these figures, it can be seen that both ε′ and ε″ increase with increasing PC content in the blends. Also, it is notable that the curves relating ε″ and the frequency are broader than one process indicating more than one relaxation mechanism. These mechanisms ascribe the rotation of the

TABLE III
ε′, T_g, and η_i Values of PC/PS Blends with Different Polyester Contents

PC content	ε′ at 100 Hz	T_g (°C)	η_i
0% polyester			
100	3.33	140	0.48
75	3.07	126	0.42
50	2.94	105	0.55
25	2.87	101	0.90
0	2.84	98	1.73
5% polyester			
100	3.396	141	0.49
75	3.36	131	0.85
50	3.21	117	1.21
25	3.05	108	1.56
0	2.96	101	1.92
10% polyester			
100	3.54	142	0.53
75	3.45	122	1.11
50	3.20	118	1.50
25	2.98	108	1.78
0	2.98	103	2.17
20% polyester			
100	3.54	144	0.61
75	3.45	136	1.69
50	3.40	125	1.88
25	3.20	115	2.01
0	2.98	105	2.38

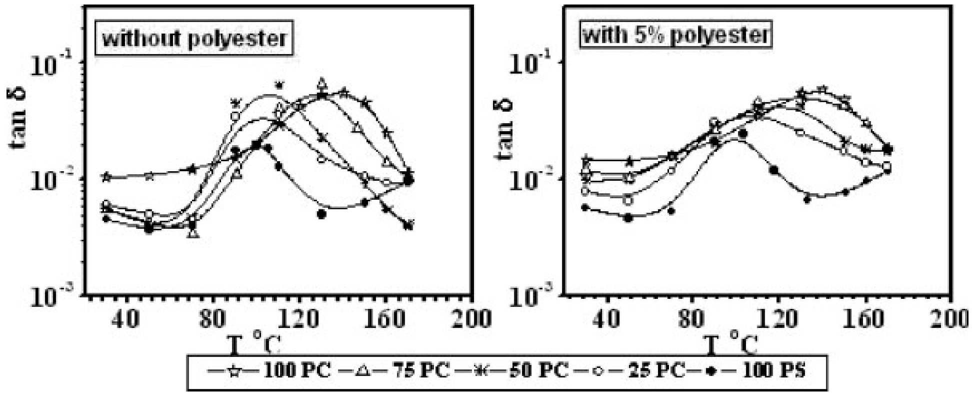


Figure 13 Variation of tan δ versus the temperature (*T*) for the PC/PS blends at 100 Hz without and with 5% polyester.

main chain and its related motions in addition to the Maxwell–Wagner effect at the lower frequency range.

Dielectric measurements

T_g. The variation of tan δ and the temperature at a frequency of 100 Hz for the blends under investigation before and after the addition of different ratios of polyester was measured, from which the T_g values were determined; they are listed in Table III.

Examples of these measurements are illustrated in Figure 13 for PC/PS blends without and with 5% polyester. Figure 14 represents the variation of T_g versus the PC content in PC/PS blends without and with 5% polyester. From this figure, it is notable that T_g for blends free from polyester deviate from the line connecting the two individuals, whereas those containing 5% polyester lie on the line connecting the values of T_g of both PC and PS. This result indicates that the addition of 5% polyester or more improves to some extent the degree of compatibility of such blends.

ε′. The values of ε′ at a frequency of 100 Hz are listed in Table III. Examples of these measurements are illustrated in Figure 13 for PC/PS blends without and with 5% polyester. From this figure, it is clear that the experimental values of ε′ fit well the solid lines of both individuals (PC and PS) after the addition of 5% polyester. This result supports those detected by T_g.

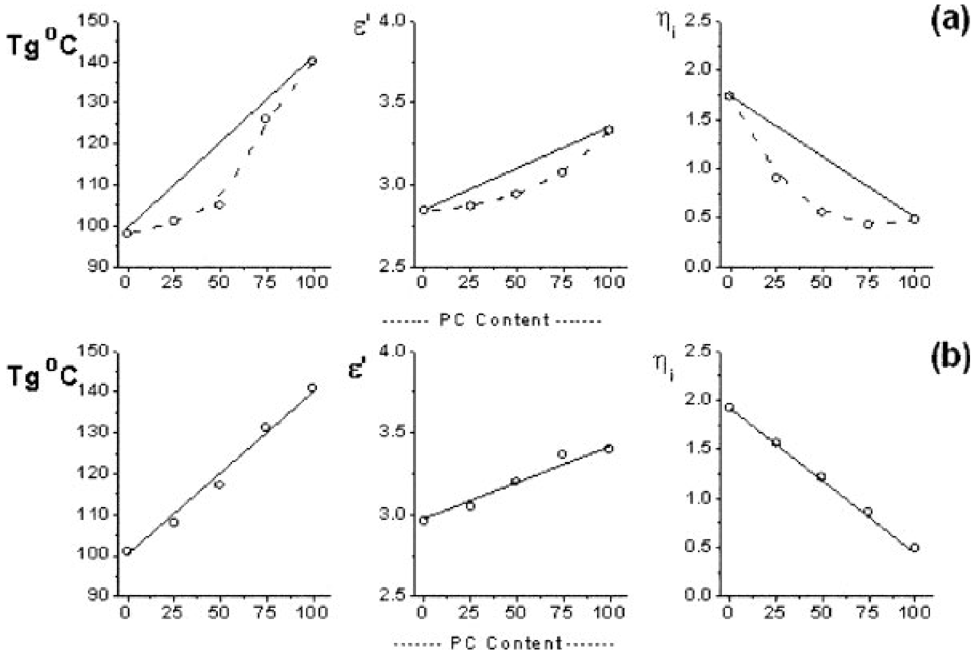


Figure 14 Variation of T_g, ε′, and η_i versus the PC content in the PC/PS blends (a) without and (b) with 5% polyester.

Viscosity measurements

The viscosity technique mentioned previously was used to confirm the two aforementioned methods. The values of η_i were obtained and are listed in Table III. The plots of η_i versus the PC contents for blends free from polyester and those containing 5% polyester are illustrated graphically in Figure 13. A straight line with a correlation factor of 0.999 was obtained after the addition of 5% polyester, and this supports the results obtained by the dielectric measurements (both T_g and ε').

CONCLUSIONS

- A polyester resin was prepared to be used as a compatibilizing agent for PMMA/PC, PMMA/PS, and PC/PS blends. The identification of the polyester was carried out with IR and ^{1}H-NMR.
- The dielectric data on frequency domains revealed more than one relaxation mechanism ascribed to the Maxwell–Wagner effect, the main chain, and its related motions.
- The compatibility was determined through dielectric (ε' and tan δ) and viscosity measurements, which are considered to be good tools for the investigation of the overall degree of compatibility of PMMA/PC, PMMA/PS, and PC/PS blends.
- The addition of 10% polyester to PMMA/PC, 20% polyester to PMMA/PS, and 5% polyester to PC/PS could improve to some extent the compatibility of such blends.

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