

Phase Behavior and Mechanical Properties of Injection Molded Poly(Ethylene Terephthalate)/Polyarylate Blends

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SYNOPSIS

Directly injection molded blends, composed of poly(ethylene terephthalate) and a polyarylate of bisphenol A, and isophthalic/terephthalic acids have been studied. Both their phase behavior and their mechanical properties after this blending method have been determined. After injection molding, the poly(ethylene terephthalate)/polyarylate blends show a single glass transition by differential scanning calorimetry, although the transition peaks of the blends widen when observed by dynamic mechanical analysis. The calorimetric results show a hindered crystallization of poly(ethylene terephthalate), due to the presence of polyarylate and the occurrence of interchange reactions. The mechanical behavior observed shows an improvement in the small-strain properties of the blends with respect to those of the pure components. The break and impact properties of the blends show negative deviations with respect to linearity. These deviations are less important when high polyarylate content exists in the blends.

INTRODUCTION

Polymer blends are an important route for the development of new polymeric materials. The importance has been demonstrated in recent years by the intensive research carried out on the subject, the increasing amount of articles, patents, and books that are continuously published, and the introduction of polymer blends for practical uses.

Among the different types of polymer blends, those containing polyesters have been extensively studied in the literature, principally with bisphenol A polycarbonate as a second component. Thus, blends of this polycarbonate, with engineering polyesters such as poly(ethylene terephthalate) (PET)¹⁻¹² and poly(butylene terephthalate) (PBT),¹³⁻²² as well as with other polyesters and copolyesters, have been studied.

The copolyester, composed of bisphenol A and 50/50 isophthalic/terephthalic acids (polyarylate, PAr), is an amorphous polymer, similar to bisphenol

A polycarbonate, which has been found to be miscible with PBT^{23,24} and partially miscible with PET.²⁵⁻²⁷ In the PET blends, and from the glass transition temperatures determined by differential scanning calorimetry and dynamic-mechanical analysis, it appears that blends with PAr contents equal to or higher than 80% are composed of a single mixed amorphous phase,^{26,27} whereas those with PAr content lower than the above mentioned contents are composed of two phases, a practically pure PET phase and a second PAr-rich phase.²⁵⁻²⁷ The melt rheology of these blends has recently been studied.²⁸

It has also been demonstrated that blends composed of PAr and PET may give rise to interchange reactions when they are maintained in the melt state.^{25-27,29} The products obtained from interchange reactions are initially block copolymers, which are progressively transformed in random copolymers. As a consequence of the products obtained, the mixtures are progressively homogenized until they finally give rise to single-phase products. Another consequence of interchange reactions is the decrease in the ability of PET to crystallize and the retention of less perfect PET crystallites, due to the decrease in the crystallizable segment length distribution of this polymer.³⁰

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Very recently,³¹ blends of PET with polyarylate and bisphenol A polycarbonate have been studied as possible products for blow molding with a higher glass transition temperature than pure PET.

The works concerning miscibility that have been mentioned were carried out after an effective full mixing, and the works on interchange reactions show that they may be controlled³² by means of the mixing temperature and time. Thus, the next step in the development of these blends would be to determine their properties when they are directly mixed in a molding process, such as injection molding. This study should be carried out by mixing and processing directly, in an injection machine under normal processing conditions, pellets of PAR and PET to determine (a) whether a full mixing is achieved and, mainly, (b) the extent, if any, of the interchange reactions that may take place. The static mechanical and impact properties of the blends obtained by the proposed molding method, to our knowledge, have not been reported. The level of interchange reactions and its influence on the phase behavior and properties of the material will help us to determine the processing conditions, which will produce the desired reaction level.

These are the reasons for our study, in this work, of the mixing level and the phase behavior of PET/PAR blends that are directly injection molded from dry mixed pellets of both components. In the second part of the work, the mechanical properties of the blends have also been studied by means of the tensile and impact tests.

EXPERIMENTAL

The polymers used in this work were polyarylate (PAR) Arilef U-1060 (Solvay) and poly(ethylene terephthalate) (PET) (Brilen S. A., Barbastro, Spain). The spectroscopic analysis of PAR indicates that it is a copolymer of bisphenol A/isophthalic acid/terephthalic acid (2 : 1 : 1), although the NMR spectrum indicates the presence of small amounts of unknown products. Thus, this polyarylate may be considered to have the above-mentioned composition. The mol wt of polyarylate was determined by viscometry in THF at 25°C. It is $M_v = 34,900$. The poly(ethylene terephthalate) had mol wt $M_n = 25,000$, measured by viscometry in *o*-chlorophenol at 35°C.

Both polymers were dried *in vacuo* at 80°C for a period of 14 h before processing. With these drying conditions, residual moisture contents of 0.07% and 0.03% were obtained for PET and PAR, respectively.

Relative measurements of the melt viscosity of the blends were obtained from the torque during kneading in a Brabender Plasticorder at 300°C, at a mixing blade speed of 30 r.p.m.

Dry PET/PAR mixtures in 100/0, 75/25, 60/40, 50/50, 40/60, 25/75, and 0/100 compositions were injection molded, using a Battenfeld BA 230E injection molding machine. The barrel temperature was 300°C, and the mold temperature was 15°C. A screw speed of 150 r.p.m. during plasticization, and an injection speed of 4.5 cm/s, were used. Tensile (ASTM D638, type IV) and impact (ASTM D256) specimens were obtained by injection molding.

¹H NMR analysis of the mixtures after injection molding was carried out to determine the existence and, in the event of this existence, the level of interchange reactions. A Varian VXR 300 spectrometer was used. The samples were dissolved in a deuterated chloroform/deuterated trifluoroacetic acid mixture and tetramethyl silane was the internal reference standard.

DSC measurements were performed with a Perkin-Elmer DSC-2 calorimeter, equipped with a Perkin-Elmer TADS system. A heating rate of 20 K/min was used and a nitrogen flow was maintained through the sample and reference chambers. The thermal transitions of the blends were determined in the usual way. The temperature and the enthalpy were calibrated with reference to an indium standard.

Dynamic-mechanical analysis was carried out on a DMTA from Polymer Laboratories, which provided the storage (E') and loss (E'') moduli and the loss tangent ($\tan \delta$). A heating rate of 4°C/min was used at a frequency of 1 Hz.

Vicat softening temperatures were determined according to ASTM D1525 (50°C/h and 1000 g load). Density measurements were carried out at 23°C in a gradient density column using sodium nitrate solutions.

Tensile tests on PET/PAR blends were carried out using an Instron 4301 tensile tester. A crosshead speed of 10 mm/min was used. The temperature was 23°C and several mechanical properties (Young's modulus, E ; nominal yield stress, σ_y ; nominal break stress, σ_b and ductility, as measured from the nominal deformation at break, ϵ_b) were determined from the force-displacement curves.

Izod impact tests were carried out on notched specimens using a CEAST pendulum, equipped with an AFS/MK3 modulus. The notches (depth = 2.54 mm) were machined after injection molding. A minimum of 8 specimens were tested for each determination in both tensile and impact tests.

RESULTS AND DISCUSSION

Phase Behavior

Preliminary work on the viscosity of the PET/PAr blends, which is of interest and is not usually known in injection molding, and an indication of the miscibility level of the PET/PAr blends in the melt state were obtained by means of the torque-composition relationship of the blends. This relationship is shown in Figure 1 and was obtained by kneading PET and PAr in a Brabender Plasticorder. The torque values correspond to the steady-state obtained after the minimum time, during which melting, plasticization, and full mixing are considered to take place. Thus, the represented torque values correspond to the minimum-reacted PET/PAr mixtures.

As is known,³³ the logarithm of the torque required to turn the Brabender is related to the melt viscosity of the blend, although the relation is somewhat complex. In our case, the method of measuring melt viscosity is especially suitable because of the impossibility of using other methods, such as MFI (Melt Flow Index) measurements, due to the different melt viscosities between both blend components. The torque values observed in Figure 1 indicate that the viscosity of the blends is clearly lower than that which corresponds to the linear relationship between the values of the pure components. Negative deviations in the torque-composition relationship, with respect to linearity, have been suggested to be indicative of a poor compatibility between the blend components in the melt,^{32,34} although this relation is not clear at the present time.^{33,35}

Figure 2 shows the thermal transitions of the injection molded PET/PAr blends, obtained by DSC

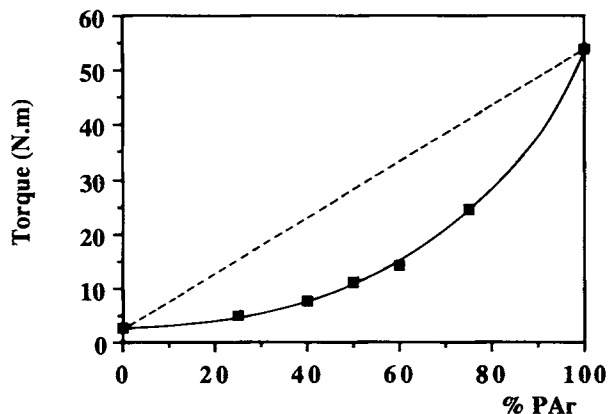


Figure 1 Torque-composition relationship for PET/PAr blends.

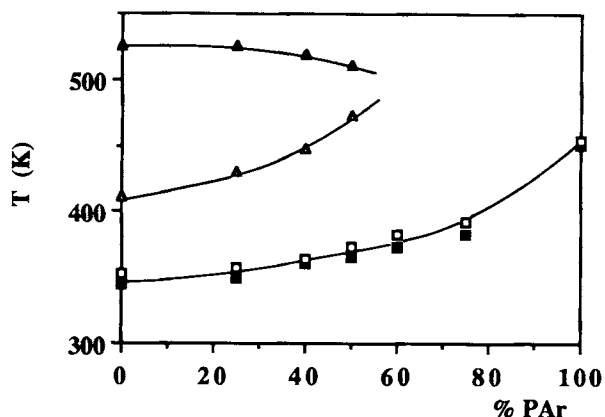


Figure 2 Thermal transitions of PET/PAr blends. (■) T_g (DSC), (□) T_g (DMTA), (△) T_c , and (▲) T_m .

and DMTA, as a function of composition. As can be seen, one glass transition, which is intermediate between those of the pure components, is observed in all the compositions studied. The T_g values increase as the PAr content in the blends increases, and the values are slightly higher when they are determined by DMTA, as is usually observed.

These results will be discussed with reference to previous studies on PET/PAr blends, in which partial miscibility between the blend components was observed. Thus, Kimura et al.²⁵ and Eguiazábal et al.²⁷ have observed a single T_g in quenched PET/PAr blends for compositions with PET contents of 30% or more. This T_g corresponds to a practically pure PET phase. However, a second T_g should appear at higher temperatures, corresponding to a PAr-rich phase. This T_g is obscured by the crystallization exotherm of PET in quenched samples. In fact, the second T_g is observed by Kimura et al.²⁵ in annealed PET/PAr blends. In our case, a single T_g also appears but, as can be observed in Figure 2, it is intermediate between those of the pure components. These data indicate, in spite of the crystallization of PET, the presence of a single phase in the blends, because, if two phases existed, a PAr-rich phase would appear at high PAr contents of the blends and the phase does not appear. This indicates that an effective mixing, at least at the level that can be detected by DSC, was achieved in the injection machine.

Robeson²⁶ observed two glass transitions by dynamic-mechanical analysis in PET/PAr blends with PET contents equal to or higher than 30%, but a single T_g for blends with a PET content lower than 30%. Eguiazábal et al.²⁷ also observed a single glass transition and no crystallization exotherms in 20/80 and 10/90 PET/PAr blends. Thus, these re-

sults indicate that blends with PET contents of 20% or less are miscible, and that PET-rich and PAR-rich phases are present at higher PET contents.

Our DMTA results, as well as our DSC results, show a single T_g in all compositions. However, the transition region in the DMTA plots is wider for the blends than for the pure components, as can be observed in Figure 3. This indicates the presence of concentration gradients in the material, but they are not great enough to be considered macroscopically as multiphasic. At high PET contents, and after the T_g corresponding to the PET-rich phase, the crystallization of PET obscured any other possible T_g .

The results of phase behavior and crystallization or melting peaks by DSC of the PET/PAr blends, obtained in this work by injection molding at 300°C, coincide appreciably with those of Robeson, which were obtained by extrusion and later by compression molding at 265–270°C. However, the usually more sensitive DMTA data, although similar at high PAr contents in both works, disagree at PAr contents of 60% or less. This is due to the presence at these PAr contents, and after the molding conditions of this work, of single glass transitions, although they are wide. These single T_g s prove the existence of interchange reactions after direct injection molding. These reactions may take place in these blends^{25–27,29} and lead to a homogenization of the blends, as has been already mentioned.

The occurrence of interchange reactions after injection molding in the selected processing conditions is proved by means of the ¹H NMR spectra, which appear in Figure 4. These spectra may be commented on with reference to the recent work of Valero et al.³⁶ Thus, the interchange reactions are demonstrated principally by the appearance of new signals in the aromatic region of the NMR spectra. These signals appear between the signal centered at $\delta = 8.16$, which corresponds to the terephthalate protons of PET, and the one centered at $\delta = 8.41$, corresponding to the terephthalate protons of PAr. Valero et al.³⁶ attribute the new signals to the aromatic protons of the terephthalate unit, asymmetrically substituted as a consequence of the transesterification reaction.

From the intensities of the new signals as a function of the blend composition, it appears that interchange reactions take place to a reduced extent, but the greater the PAr content in the blends is, the more important the reactions are.

Some effect of the different moisture levels in the blend components on the extent of interchange reactions at different blend compositions is normally expected; however, it seems to be negligible in this case, because the greater level of interchange reactions appears at high PAr contents, that is, at lower moisture contents. Thus, even though the conditions for reactions in the injection machine were not pursued, interchange reactions take place. These re-

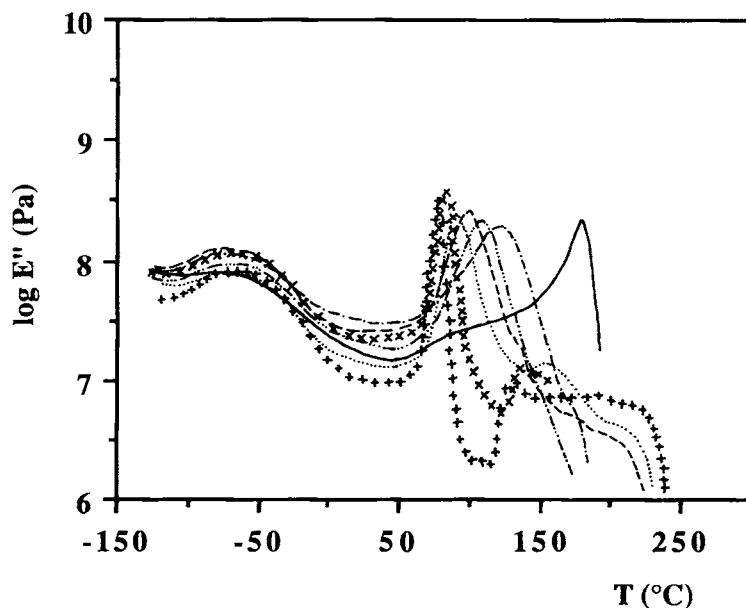


Figure 3 Dynamic mechanical spectra of PET/PAr blends. (—) 0/100, (- - - - -) 25/75, (- · - · - · -) 40/60, (- · - - -) 50/50, (· · · · ·) 60/40, (××××) 75/25, and (+++++) 100/0.

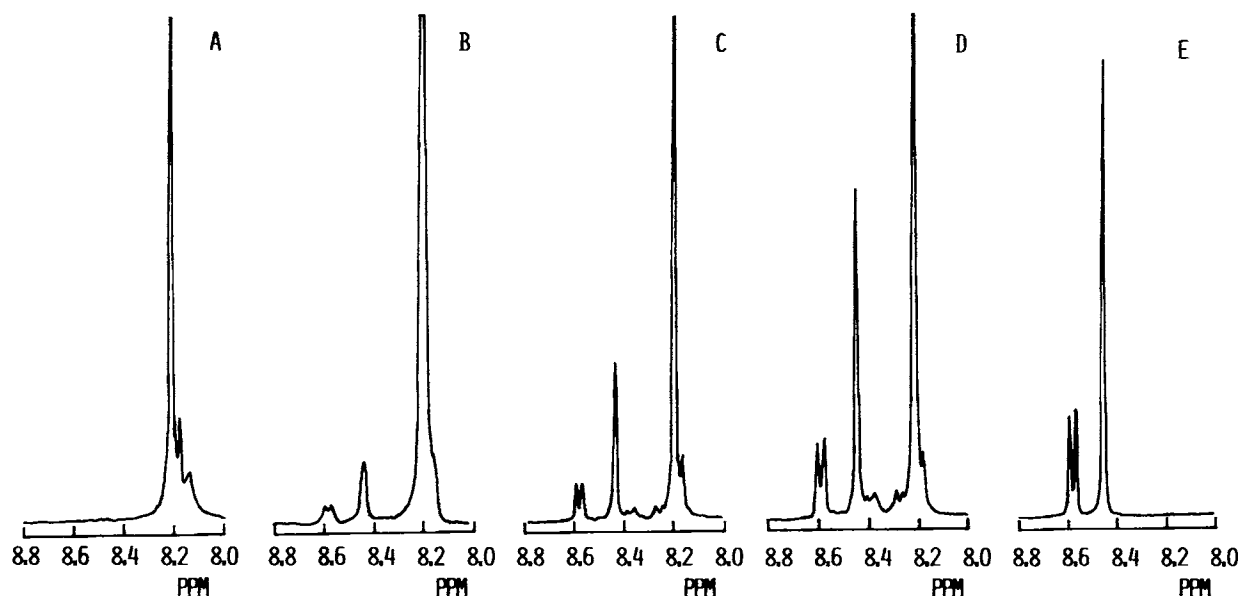


Figure 4 ^1H NMR spectra of injection molded PET/PAr blends. (A) PET, (B) PET/PAr (75/25), (C) PET/PAr (50/50), (D) PET/PAr (25/75), and (E) PAr.

actions produce, despite their small extent, a partial compatibilization of the blend to the point where they produce a single wide T_g .

The variation of the glass transition temperatures of the blends has, as a consequence, and as can be observed in Figure 5, increased the Vicat softening temperature as the PAr content in the blends increases.

The second feature of the calorimetric scans is the crystallization exotherm, which appears for blends with PET contents equal to or higher than 50%. The temperatures, corresponding to the minima of the exotherms, are represented in Figure 2. These exotherms appear to be a result of the low mold temperature and the fact that PET does not

crystallize to its equilibrium level during cooling in the mold. Thus, crystallization takes place during heating in the DSC scan. As is observed in Figure 2, the T_c of PET increases markedly as the PAr content in the blend increases. At the same time, a broadening of the crystallization peak is observed. Both results are due to the presence of PAr and to the interchange reactions, which make the crystallization of PET difficult. The effect of PAr on the crystallization temperature of PET is similar to that observed by Robeson²⁶ and different to that observed by Eguiazábal et al.²⁷ Different degrees of interchange reactions obtained during the preparation of the blends, and the different thermal treatments applied in each case, may be responsible for the different behaviors observed. The crystallization heats, measured from the areas under the corresponding peaks, decrease as the PAr content in the blends increases, as can be observed in Figure 6.

Finally, the DSC scans of the injection molded PET/PAr blends show a melting endotherm of PET when the content of the semicrystalline polymer is equal to or higher than 50%. The melting temperatures are shown in Figure 2, and clearly decrease as the PAr content in the blends increases. The decrease observed from 100% to 50% PET in the blend is 16 K. This decrease is greater than that which was expected, taking into account the partial miscibility of the blends. In fact, previous results indicate a practically constant melting temperature of PET²⁷ or a decrease smaller than that observed in

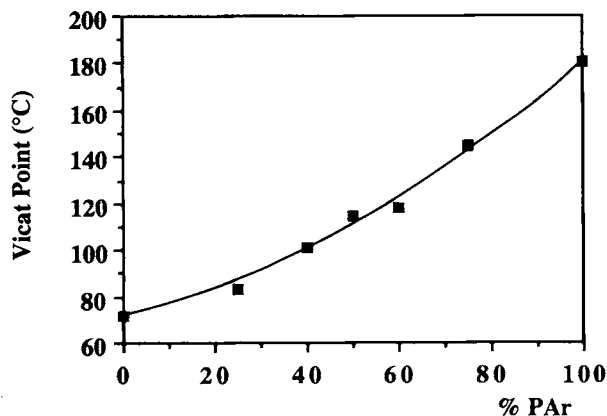


Figure 5 Vicat softening points of PET/PAr blends.

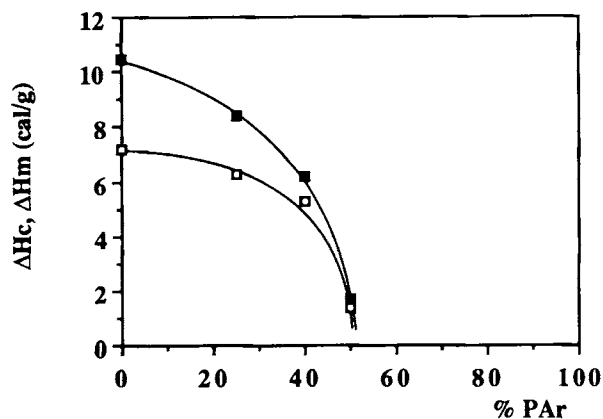


Figure 6 Crystallization (□) and melting (■) heats of PET/PAr blends.

this work,²⁶ depending on the blending method and on the thermal treatment. This decrease in the melting temperature is further evidence of the additional presence of PAr in the vicinity of the PET molecule, apart from that produced by partial miscibility. This additional presence and its consequence, the decrease in the melting temperature of PET, are a consequence of the interchange reactions that were demonstrated to take place in the present processing conditions.

The melting heats of PET in the blends are also shown in Figure 6. It is observed that the melting heat is, in all cases, greater than the crystallization heat. This indicates that some fraction of PET crystallizes during cooling in the mold. The greater the PAr content in the blends, the smaller the difference between ΔH_m and ΔH_c . This is due to the fact that, as we have mentioned, the crystallization of PET is hindered by the presence of PAr and the subsequent interchange reactions. Thus, the fraction of PET that crystallizes during cooling in the injection mold is smaller as PAr content in the blend becomes greater.

At PAr contents of 60% and more, no crystallization or melting peaks of PET appear.

In Figure 7, we show the density values obtained for the different compositions of PET/PAr blends. As can be observed, an approximately linear relationship is obtained. These results are different from those obtained by Robeson,²⁶ who observed positive deviations from linearity in the range of miscibility, probably due to the different thermal treatments, and, as a consequence, observed crystallinity levels of PET in the blends in both cases. These different density-composition plots in both works show that caution must be taken when, in the case of one of the blend components being crystalline, the

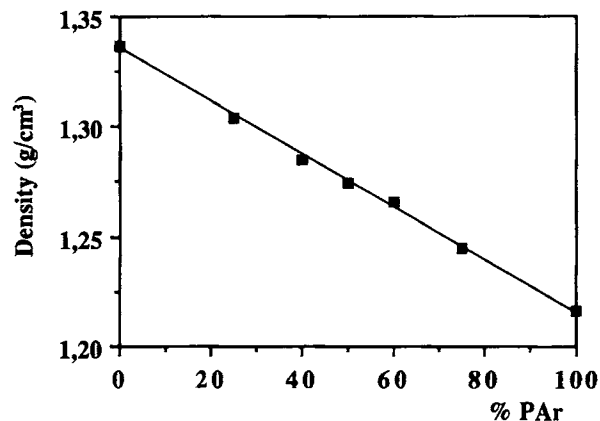


Figure 7 Density-composition relationship for PET/PAr blends.

relationship³⁷ between positive/negative deviations of density from linearity, and the corresponding existence/inexistence of strong interactions between the blend components, is accepted.

Mechanical Properties

Figure 8 shows the variation of Young's modulus of PET/PAr blends as a function of composition. As can be observed, the experimental values show a clear positive deviation from linearity, that is, a synergistic behavior. Moreover, all the E values obtained for the blends are higher than those obtained for any of the two components of the blend. This outstanding behavior is a surprising result because, although synergisms in the moduli are common, even in immiscible blends, and sometimes some compositions give moduli higher than those of the components, to our knowledge this is the first time that the complete composition range shows moduli higher than that of any of the components.

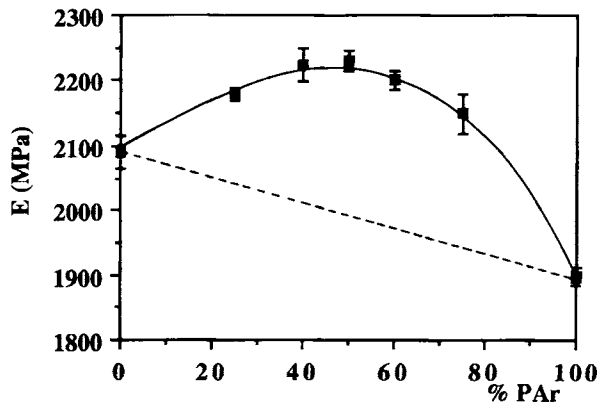


Figure 8 Young's modulus of PET/PAr blends.

In the case of synergisms, which do not have a value higher than that of the highest of the two components, it may be supposed that the component of Young's highest modulus contributes to a greater extent than that which corresponds to composition. An example of this is when the matrix, due to stress or strain concentrations, may give rise to increased local contributions to the overall stress with respect to that which corresponds to the composition of the blend. However, this kind of synergism is difficult to understand. This is even more surprising, given that the modulus increase is around 10% and that no densification has been seen from density measurements.

Synergistic behaviors in Young's modulus-composition relations in polymer blends have been explained in some cases³⁷ as a consequence of the blend densification, due to interactions between the components. In PET/PAr blends, however, as has already been mentioned, no densification has been observed in our work, and only in PAr-rich blends in another work.²⁶ Thus, this effect, as well as crystallinity, which was not above linearity, should be discarded as causes of the synergistic behavior of Young's modulus.

Another important low-strain property, yield stress, also shows a behavior characterized by a positive deviation from linearity when it is represented against the blend composition, as can be observed in Figure 9. The greater positive deviation is found for the 25/75 blend, that is, for the compositional region at which the greater miscibility exists in the blends. These results are in good agreement with those obtained for Young's modulus, and also demonstrate a good adhesion between the blend components.

The positive effect of mixing PET and PAr on the low-strain properties is not maintained when

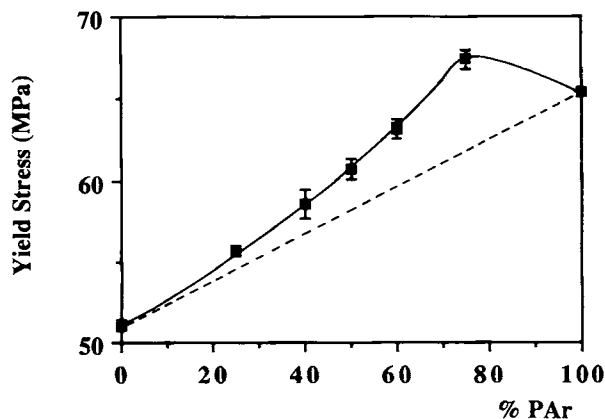


Figure 9 Yield stress of PET/PAr blends.

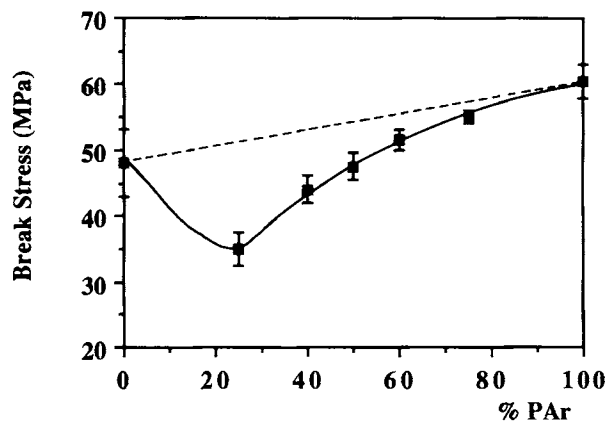


Figure 10 Break stress of PET/PAr blends.

the break properties are considered. In Figure 10, we show the break stress of the blends. As can be observed, all the experimental values are lower than those corresponding to linearity with a clear minimum for the 75/25 PET/PAr composition; the values, however, are quite close to linearity.

The effect of mixing on the break strain of the blends is even more important than that observed in the break stress, as is usual, and as can be observed in Figure 11. A dramatic reduction of this property is observed in the mixtures with respect to linearity. In the case of blends with a PAr content of 25% or more, this gives rise to the ϵ_b values for the blends to be in all cases approximately equal or slightly higher than those obtained for pure PAr. This behavior, however, is not that which corresponds to an immiscible blend. The deformation at break of the worse composition is approximately 13%; yielding took place in all the blends.

The extreme influence of incompatibility in the deformation at break of polymer blends³⁸ is well known. Incompatibility gives rise to deformations

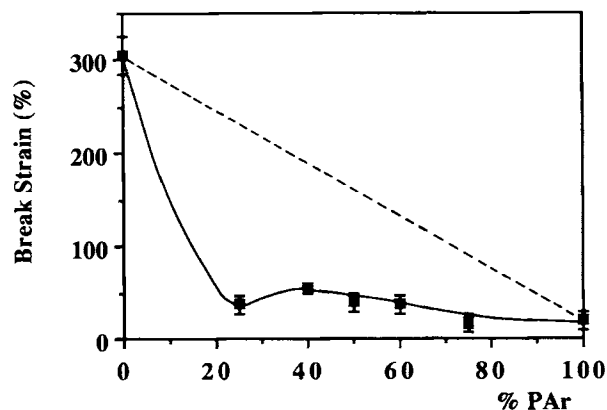


Figure 11 Break strain of PET/PAr blends.

at break on the order of 1%. Thus, the ductility level of the PET/PAr blends must be a consequence of the combined positive effects of partial miscibility and of the transesterification reactions that may have a positive effect on ductility.³²

Finally, the impact strength data are shown in Table I. It is observed that a great difference exists between the impact strength of the two blend components. For the blends, low impact strength values, lower than those of pure PET, are obtained except in the PET/PAr (25/75) blend. Thus, the impact strength is more greatly affected by mixing than the deformation at break corresponding to the decreases in ductility that are usually accompanied by the high strain rate, characteristic of the impact test. The greatest negative effect is observed again at high and medium PET contents in the blends.

The results obtained for the different blend compositions in break and impact properties may be explained on the basis of the miscibility level characteristic of each composition and of the interchange reactions that take place in the blend. Thus, from NMR, it appears that the greater reaction level is obtained at high PAr contents and, in the case of nonreacted blends, it was seen that maximum miscibility takes place also at high PAr contents.^{26,27} These experimental evidences are in agreement with the fact that, at these high PAr content compositions, the break properties show the smaller deviation from linearity. On the contrary, the more important negative deviations are obtained in the PET/PAr (75/25) blend.

The results obtained in this work show some interesting aspects that may be of practical application in the cases of both low and high PAr contents in the blends. In the case of blends with medium and high PET contents, although the deterioration of the break properties is an important disadvantage for the practical application of these blends, they also show an increase in the small-strain properties with respect to pure PET, principally in the case of

Young's modulus. Moreover, the principal advantages, with respect to pure PET, are the increase in the T_g and in the Vicat softening point, both of which are low in PET, and which constitute one of pure PET's disadvantages. A further advantage is the hindrance of PET crystallization, which is caused by the presence of PAr and by the reactions produced during injection molding. This offers a method for obtaining PET-based amorphous products with improved thermal resistance. This balance of properties may be changed, and tailored blends may be obtained by changing both the PAr content of the blend and the injection parameters.

At high PAr contents in the blends, the blends are amorphous. The most negative effect of mixing at this compositional level is the decrease in the glass transition temperature and in the Vicat softening point, with respect to those of pure PAr. However, PAr is mainly used as an engineering material, because of its good mechanical properties, so that some positive aspects need to be considered. First, the small-strain properties of the blends increase with respect to those of pure PAr and the break properties are practically unaffected by mixing. The exception to this generally positive behavior is the impact strength, which decreases markedly from pure PAr to the PET/PAr (25/75) composition, possibly due to the very small impact strength of PET. This ensemble of properties, plus the sharp decrease in the melt viscosity of the blend with respect to that of pure PAr, occur at the same time. Thus, this blend must be processed more readily than the pure amorphous polymer, giving an additional advantage for its possible use.

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Table I Impact Strength of PET/PAr Blends

Composition (PET/PAr)	Impact Strength (J/m)
100/0	32
75/25	19
60/40	18
50/50	15
40/60	15
25/75	176
0/100	459

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