

Control of Interchange Reactions of Polycarbonate / Polyarylate Blends and Their Influence on Physical Behavior

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Synopsis

Blends of polycarbonate of bisphenol A (PC) and a polyarylate of bisphenol A (PAr) are susceptible to showing interchange reactions in the melt state. The control of these reactions was carried out by means of the observation of the torque required to turn the Brabender and of its increase against the time due to copolymer formation in the processing equipment. Based on this variation and on glass transition temperature (T_g) measurements, the possibility of an exchange reaction in two steps in this blend was suggested. T_g measurements in melt- and solvent-cast blends also showed that this mixture is immiscible at all compositions and that, by copolymer evolution, a single T_g intermediate between those of the individual constituents can be found in all compositions. The influence of immiscibility on the mechanical properties of the blends was shown by the appearance of a minimum in large-strain properties at about 25% PAr. The behavior of the transesterified blends was very different showing a clear improvement of the tensile properties compared with those of the corresponding blends.

INTRODUCTION

The commercial application of polymer blends is becoming more and more widespread. The reason for its economic importance is that a polymer mixture system offers a way of meeting new market applications with a minimum development cost. This way is not well known; thus, the properties of polymer blends depend not only on the miscibility level of the components, but also on the processing conditions, especially in the case of immiscible mixtures. Thus, adequate processing conditions can lead to a useful compatible blend from two immiscible polymers.

If mixing has to take place in a melted state, the intimate control of this state during the mixing together of the components provides a clear opportunity for the development of chemical reactions between both polymer reactive groups. In this way, the existence of these exchange reactions between some polymer pairs¹⁻²⁰ is well known, mainly between polyesters.¹⁻¹⁸

Transesterification makes it possible to obtain copolymers with different levels of randomness and composition. This level is a function of the temperature and residence time in the melt state, as well as a function of the percentage of each mixed polymer.^{3,5,7,9-11,14,16,20,21} However, except in some works,^{10,19} there have been no studies of the control of these reactions during processing.

The control of interchange reactions in those polymer blends that can produce these reactions should allow us to obtain alloys which are more homogeneous than the corresponding physical mixtures, mainly in cases of immiscibility. This would more than likely mean that the properties of the mixture, and particularly the mechanical behavior, would be better than those of the physical blend; however, there is little information on the bibliography of these variations of properties.

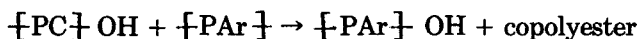
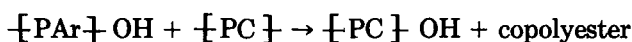
During the development of transesterification reactions we may expect a change to occur in the melt viscosity due to the newly formed structure, and this variation should show an increase in this property until both blend components have fully reacted. This viscosity variation must produce the corresponding change in the torque required to turn the Brabender.

The purpose of this paper is to investigate the influence of transesterification reactions between a polyarylate (PAR) and polycarbonate of bisphenol A (PC) on the mechanical properties and on the physical state of the blend. The results will be compared with those obtained in this blend by other authors. Thus, we suggest a method of controlling these interchange reactions during processing in a Brabender Plasticorder by plotting the torque variation against residence time. This will allow us to compare the properties of the mixture before and after transesterification. An exchange reaction mechanism that analyzes the viscosity variation as a function of time and blend composition at various temperatures is also suggested.

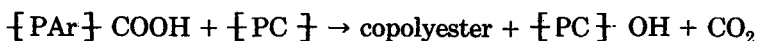
THEORY

As has been shown in some studies,¹⁻¹⁸ chemical reactions are possible between two melt-mixed polyesters or between PC and a polyester. When chains are terminated by hydroxyl or carboxyl groups, three types of interchange processes can take place.^{4,7,8,10,12,16} These reactions are:

Alcoholysis:



Acidolysis:



Direct transesterification:



Devaux et al.^{7,13-16} concluded that for the PC/poly(butylene terephthalate) (PBT) pair, the most likely mechanism of exchange reaction was the direct

ester-ester interchange. Because PAr is a polyester (similar to PBT), it can be assumed that in the PAr/PC mixture the reaction that may take place is the same, as has been shown by Kimura and Porter.³

These transesterification reactions in polyester blends lead initially to block copolymers and finally to a random copolymer,^{3-5,7,9,10,13-16,21} since all repeat bond linkages of a given structural unit are equally likely to react. The relative population of homopolymer and block copolymers depends on the moles of bonds interchanged,^{4,5,21} i.e., the properties of a blend that is transesterifying will depend on the level of the exchange reaction produced, and this should be more evident in incompatible or partially compatible polymer mixtures.

In some papers, it has been mentioned that the control of this type of chemical reactions between two polymers should be of interest from a scientific and commercial point of view. However, the control of these chemical rearrangements in a transformation process has not been achieved except in the work of Robeson and Furtek¹⁹ and Kresse.¹⁰ They showed that by means of melt-viscosity control it is possible to follow the evolution of these reactions in a processing machine such as an extruder.

On the other hand, it has been shown that miscibility between PC and polyesters is probably the result of an *n*-complex formation between the electrons of the ester carbonyl and the aromatic ring of the carbonate molecule,²²⁻²⁴ but it has also been indicated that esteric restrictions to interactions between the polymer repeat units may again be responsible for partial miscibility or, in an extreme case, for immiscibility.^{12,22-30} That is to say, the presence of strong interactions is a necessary but not sufficient condition for polymer blend miscibility. This is possibly the reason for the behavior of the PAr/PC mixture shown below.

EXPERIMENTAL

The polyarylate (PAr) used was Arylef U-100 and was provided by Solvay. It has an \bar{M}_n of 2.12×10^4 and an \bar{M}_w of 5.14×10^4 and is a copolymer of bisphenol A with a mixture of terephthalic/isophthalic acids at a proportion of 50/50. The bisphenol A polycarbonate (PC) is a commercial product produced by Bayer and known as Makrolon 2800; it has an \bar{M}_n of 1.70×10^4 and an \bar{M}_w of 3.50×10^4 . Molecular weights were measured by means of gel permeation chromatography (GPC). Neither of the melt-processed polymers showed traces of crystallinity. However, it is known that solvent-cast PC can exhibit crystallinity.

Polymer blends were prepared by melt mixing in a Brabender Plastograph using the following procedure: Pellets of the two polymers were mixed to the desired composition and dried at 120°C to ensure removal of any moisture absorbed. This mixture was transferred to the Plastograph bowl which operated at a mixing blade speed of about 5 rpm. When the bowl was full, the speed was increased to a constant value of 30 rpm and maintained until the mixing operation ended. This method was used at 250, 270, and 290°C for all blend compositions and also for the pure polymers in order to ensure the same treatment in all cases. The torque was registered vs. residence time at three different temperatures.

After the mixing operation, compression-molded sheets 0.6–0.8 mm thick were prepared from the melt-mixed blends at 250°C for 5 min and then cooled under pressure to 70°C at about 10°C/min.

Tensile test specimens (ASTM D638, type IV) were punched out from the sheets with a pneumatic die. The tensile stress–strain properties were determined on an Instron Tensile Tester using a constant crosshead speed of 0.5 mm/min (nominal strain rate of $1.3 \times 10^4 \text{ seg}^{-1}$) at $23 \pm 2^\circ\text{C}$. Yield tensile strength, elongation at break, break strength, and initial modulus were computed from the recorded load–time traces. The modulus is taken to be the initial slope of the force–deflection curve and suitable instrumental and clamp corrections were carried out before calculating it. For each property, an average for approximately 24 samples was computed.

The blends were examined by means of differential scanning calorimetry (Perkin-Elmer, DSC-2) at 20 K/min in order to establish their glass transition temperatures (T_g). Sample weights were about 10 mg and an empty sample pan was used as reference. The T_g was taken to be the point of intersection of the extrapolated low-temperature baseline with the line of maximum slope encountered during the shift.

RESULTS AND DISCUSSION

For the purpose of checking whether PAr and PC undergo transesterification during processing together, the torque required to turn the Brabender during blend mixing vs. residence time was registered. This torque is an indicator of melt viscosity, although the relation is somewhat complex.^{31–34}

If interchange reactions did not occur, at the temperatures used in processing, the melt viscosity of mixtures should remain constant or, in case of degradation, decrease vs. residence time. In order to check that the viscosity increase in blending was due to chemical reactions between mixture components, both pure polymers were processed in the same conditions as the blends. In both cases, no maximum but little decrease of torque vs. residence time was observed; this decrease was a little higher in the PAr case. This is clear evidence of the nonexistence of such reactions between the groups of each polymer.

The torque–time measurements of blends conducted in the Brabender at 270°C are illustrated in Figure 1(b). For all compositions the torque drops initially to steady; at this moment the two blend components are homogeneously mixed. If the residence time is higher, the viscosity increases progressively until it reaches a maximum value, which is attributed to a chemical interchange reaction between the functional groups of both polymers. At maximum torque, we observed that degradation set in because of a serious decrease in viscosity. This is clear evidence that transesterification is halted or, at least, that the degradation effect is higher than that which came from the chemical rearrangements between both melt polymers. Robeson and Furtak¹⁹ have shown a similar but higher increase for the miscible PBT/polyhydroxyether of bisphenol A Phenoxy blend, and, although in the PAr/PC mixture the chemical reactions that ought to take place are not the same, this maximum could be a qualitative measure of the transesterification produced.

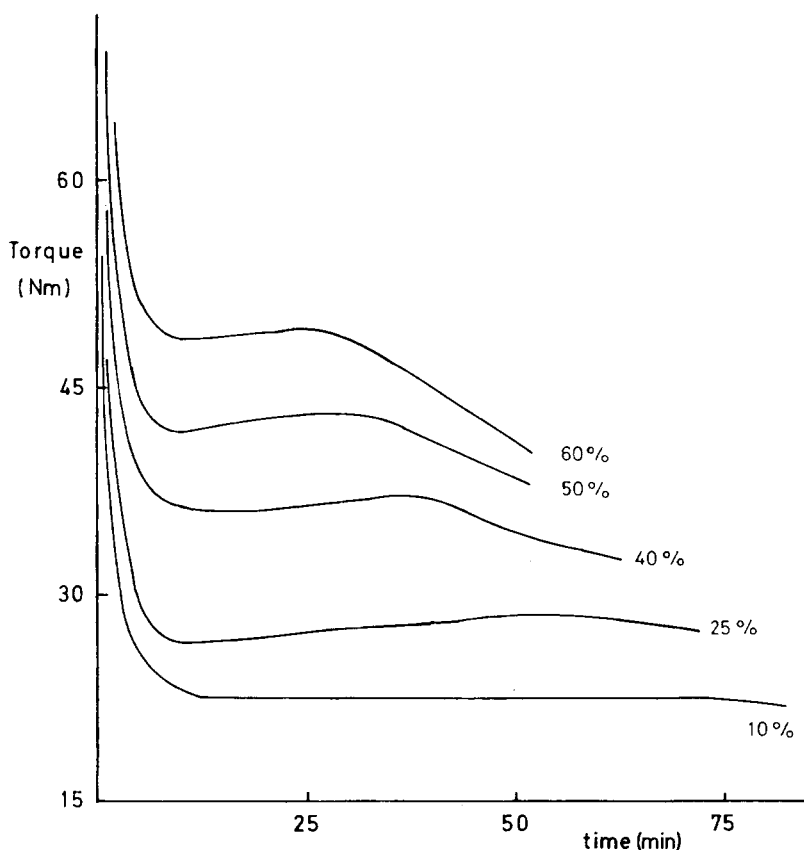


Fig. 1a

Fig. 1. Torque variations of different PAr compositions vs. residence time at (a) 250°C, (b) 270°C, and (c) 290°C.

The torque increase should not be very great because these transesterification reactions do not produce crosslinking between both polymers. This small torque increase may not be observable in other polyesters because of the degradation that can take place at the same time. Consequently, we have studied the melt behavior of the PAr/PBT and PC/PBT (and PC/PET) mixtures that are susceptible to transesterification, and such a torque increase has not been observed. Elsewhere, we have analyzed blends that are susceptible to crosslinking or to side reactions such as PAr/Phenoxy and PC/Phenoxy pairs,^{35,36} observing a torque increase similar to that in PBT/Phenoxy,¹⁹ and which is much higher than that of PAr/PC mixture.

As was expected, at 250°C the torque also shows a maximum, but at higher residence times. The torque increase varies depending on the composition and the major variation appears to occur at about 25% PAr, as shown in Figure 1(a). Similar results are observed in Figure 1(c) when the processing temperature is 290°C. Under these conditions the maximum torque appears at lower times and, as has been shown at the other temperatures, the highest increase is again observed at about 25% PAr.

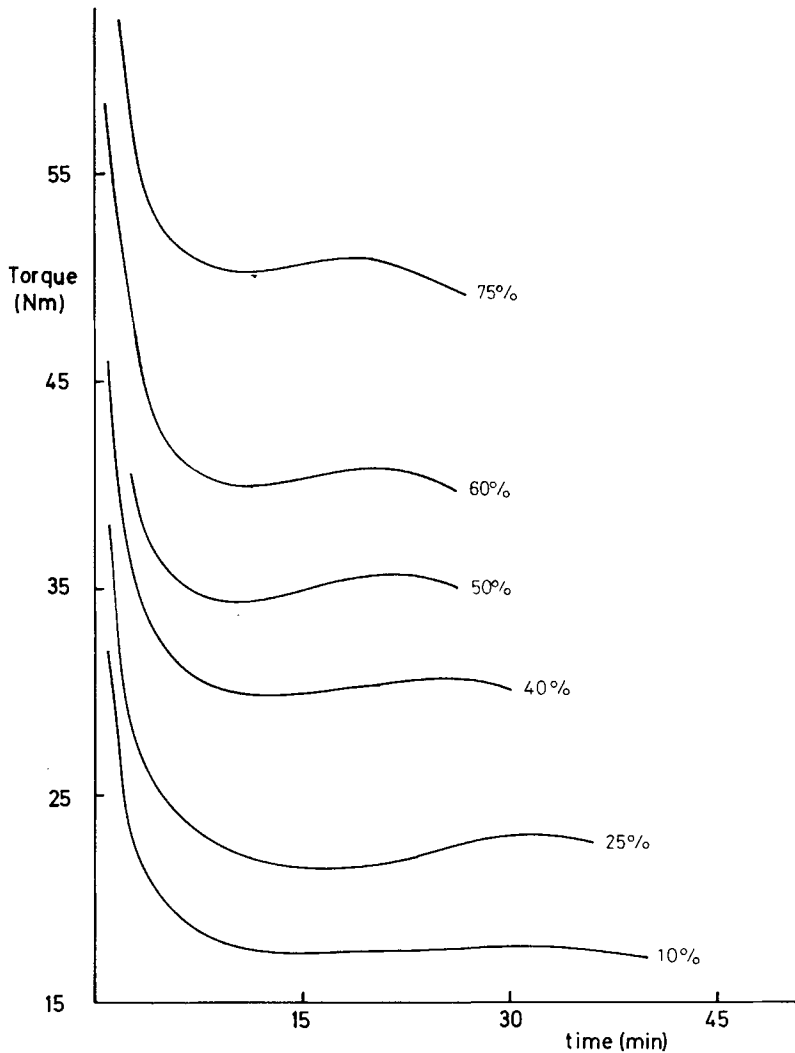


Fig. 1b

As shown in Figure 1, the lower the PAr content, the longer the maximum takes to appear and the torque change appears greater at low PAr contents. This is brought about by the higher viscosity of PAr that, in these processing conditions, causes the melt temperature to increase at the same time as the PAr content increases. As a consequence, interchange reactions are faster when the blend has a high PAr content.

If we look at Figure 2, the higher the PAr content, the closer the steadied-torque value appears to the line connecting the steadied-torque of the pure polymers. This could suggest that in these PAr-rich compositions, miscibility exists, but, as is shown below, these polymers are immiscible (or at least partially miscible) in all compositions so that this torque behavior suggests that a first transesterification step develops in the PAr-rich phase. If this exchange reaction did not exist, steadied-torque data would be below this line

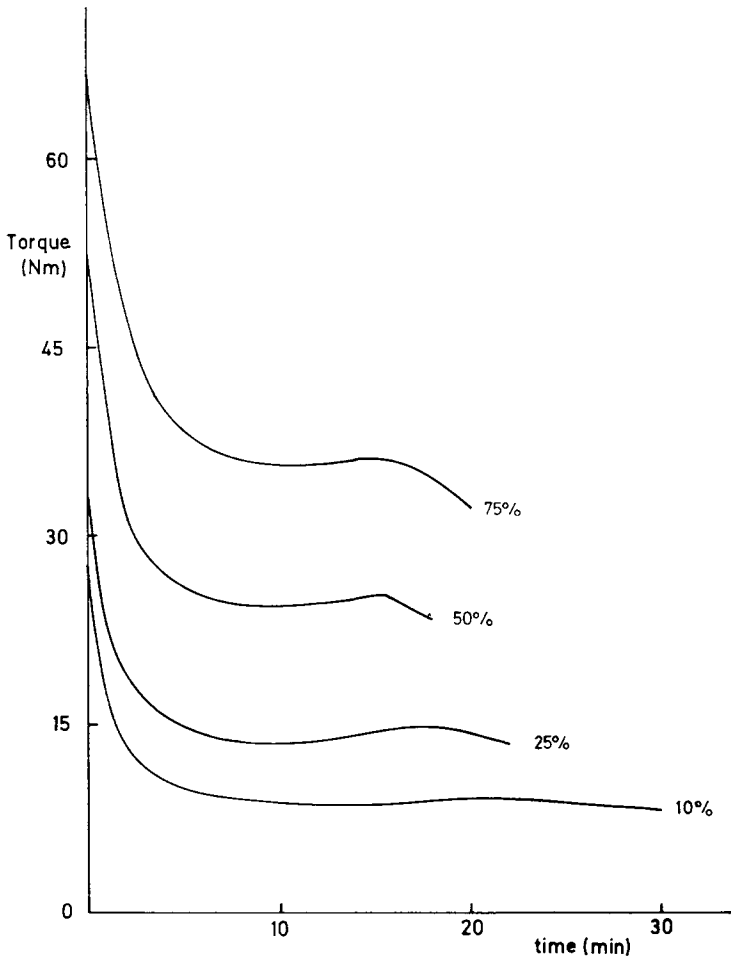


Fig. 1c

in all compositions. At low PAr contents, the amount of copolymer which can be produced in this first step would be low because of the lower melt temperatures, so that it would have lesser influence on the steadied torque, as is shown in Figure 2.

If we analyze Figures 1 and 2, it can be seen that interchange reaction evolution comprises two steps: the first step is fast, it occurs in the PAr-rich phase, and it is not observable in the torque-time plot. The second step proceeds between the two phases of the blend and produces the slope variation in the torque. As shown in Figure 1, in low PAr contents, the variation between the maximum and the steadied torque is higher. The reason for this is that the second step of the interchange reaction is more important because of the smaller amount of PAr-rich phase, i.e., because of the copolymer produced in the first step, in the overall blend.

During blend preparation in the Brabender it was observed that when the torque steadied, the transparency of mixtures varied with the temperature used. Thus, at 250°C, blends with a PAr content of more than 60% were

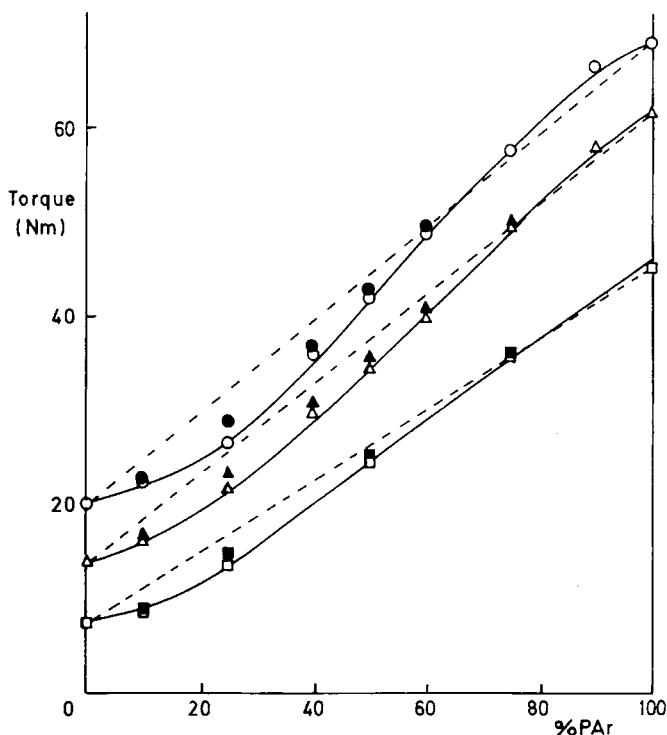


Fig. 2. Torque data of blends before transesterification (clear) and after transesterification (dark) versus mixture composition at the temperatures used: (○) 250°C; (△) 270°C; (□) 290°C.

transparent; at 270°C this limit decreased to 50% PAR, and at 290°C even this composition was observed to be transparent. Blends with a smaller PAR content appeared opaque at this temperature.

These observations may suggest that in this blend there exists a miscibility limit between 50 and 60% PAR, but transparency criteria to determine miscibility are often not valid,³⁶⁻³⁸ and, as has been observed in the T_g study of solution-cast mixtures, there are two phases in all blend compositions. In spite of this, in melt-processed blends with high PAR contents, transparency can be due, as mentioned above, to copolymer formation in the PAR-rich phase, which could act as a compatibilizing agent when it constitutes a considerable proportion of the blend, i.e., at high PAR content. Moreover, the higher melt-temperature increase at these proportions implies that transesterification (between two phases) takes place more quickly, and for this reason the higher the temperature, the lower the PAR content necessary for transparency to be observed.

At all compositions when the maximum in the torque-time curves was attained, transparency was observed in both melt and solid states. This is a clear indication of a structure change in the initially immiscible blends due to the transesterification produced, and it shows that formed copolymers were fully amorphous.

The above considerations indicate that by selecting an adequate temperature and time, copolymer formation can be controlled. At this moment, we are

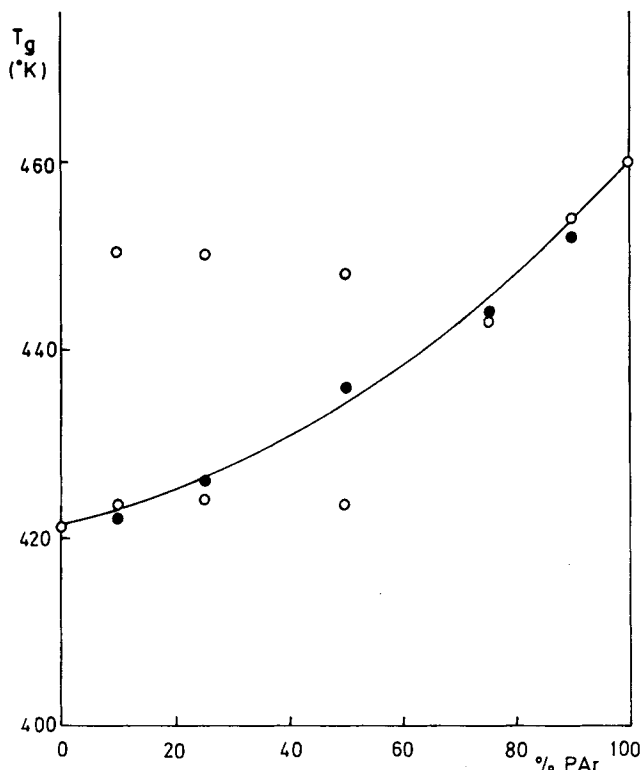


Fig. 3. The T_g 's of the mixtures before (○) and after (●) interchange reactions vs. blend content.

studying the influence of temperature and rotor speed on the control of the development of transesterification. The results will be published in the near future.

If the blends at steady torque correspond to the mixture before transesterification and the blends at maximum torque correspond to the mixture after interchange reaction, i.e., to a copolymer, the T_g results obtained by DSC should show, in case of immiscibility, two T_g 's for blends corresponding to steady torque and only one T_g for the copolymer obtained at maximum torque. Moreover, this trend should be confirmed for all blend compositions.

The results obtained for blends corresponding to steady-torque at 270°C and a rotor speed of 30 rpm are shown in Figure 3. As can be observed, in compositions lower than 75% PAr there are two T_g 's, one close to that of pure PC and the other lower than that of pure PAr, which is not easily observable at PC-rich compositions. Nevertheless, when PAr content is 75% or higher, only one T_g is observed, and this corroborates the fact that the first step of transesterification proceeds in the PAr-rich phase which has a practically constant composition in all mixtures as observed by T_g data. The higher the PAr content, the greater the weight of this PAr-rich phase with respect to the overall blend, and, consequently, more copolymer is formed, thus improving the adhesion between both phases and aiding the second step in the exchange reaction.

In Figure 3 the T_g data of the copolymers corresponding to maximum torque are also shown. At all compositions, a single T_g is observed, which is intermediate to those of two polymer constituents and is consistent with what has been shown in other studies.^{3,39,40} These T_g values can be well adapted by the Gordon-Taylor⁴¹ equation:

$$T_g = \frac{w_1 T_{g_1} + k w_2 T_{g_2}}{w_1 + k w_2}$$

where T_g is the glass transition temperature of the blend, T_{g_1} and T_{g_2} are the glass transition temperatures for the pure components, w_1 and w_2 are the corresponding weight fractions, and k is an adjusting parameter related to the degree of curvature of the T_g -blend composition diagram. If these results are compared with those obtained by extrusion in a previous study,³⁹ it can be observed that these T_g values and those of the extruded samples are very similar. This implies that the conditions used in extrusion processing were strong enough for transesterification to take place and that there is clear evidence showing that when the maximum is attained, the copolymer that corresponds to the product of the interchange reaction has been obtained. Moreover, the torque decreases after the maximum indicates that from this moment the effect of the degradation is stronger than that of the exchange reactions, if there are any. Furthermore, the lack of crystallinity traces in DSC thermograms corroborates the amorphous nature of the copolymer formed. As studied by Kimura and Porter,³ the fully transesterified copolymer corresponding to the maximum torque must have a random distribution because at this point, the interchange reaction finishes or, at least, has no importance.

To verify these data, the study carried out in a previous paper³⁹ has been repeated preparing the samples by solution casting, and analyzing them by DSC. The results are shown in Table I. Thus, when the thermograms are studied, it can be observed that at the first scan it is not possible to obtain precise conclusions due to the PC crystallinity that occlude the range of the

TABLE I
The T_g 's of the PC/PAr Mixtures after Several Thermal Histories

Thermal history	% PAr								
	100	90	75	60	50	40	25	10	0
2 min		Unclear	424	424	422	423	420	419	
at 573 K	455	446	448	447	450	449	449	Unclear	415
10 min									
at 573 K	456	447	445	436	433	430	426	419	415
Maximum torque			444		436		426	422	421
Extruded sample	460		440	434	431	427	424	418	415
T_g of Kimura and Porter ³	460		442		434		425		419

TABLE II
The T_g 's of the PC/PAr Solvent-Cast Blends after Different Delay Times in the Calorimeter

% PAr	T (K)	t (min) at T						
		2	5	10	15	30	60	90
50	523	418	419	418	421	421.5	427	430
60	523	450	450	450	447	447	Unclear	Unclear
		422	422	422	422	422	427	433
50	530	453	453	450	450	450	Unclear	
		418	420		430		430	
50	573	444	443					
		422		430	430			
60	573	450						
		422			432			
		452						

appearance of the T_g . Results from samples maintained at 573 K for 2 min showed two T_g 's at all compositions; one T_g is close to that of pure PC, and the other is lower than that of pure PAr. When these blends were kept at the same temperature for 10 min, a single T_g intermediate between those of the two polymer constituents was observed. It was similar, within experimental error, to those from samples corresponding to maximum torque, i.e., to those from transesterified blends, and likewise similar to those of blends prepared by extrusion in a previous investigation³⁹ and to those reported by Kimura and Porter³ (as shown in Table I). This is strong evidence that whatever the preparation method used, random copolymers developed because of an ester-ester interchange reaction.

When 50:50 and 60:40 PAr/PC solvent-cast samples were analyzed by DSC after maintaining them at 523, 530, and 573 K during different residence times, it was observed that, before copolymer formation, there were two T_g 's. One of these T_g 's was close to that of the pure PC, and hardly varied, and the other was lower than that of the pure PAr, and diminished somewhat at higher residence times towards the T_g of the copolymer. At longer periods of time, only one T_g was observed, and this is attributed to the copolymer formed by the ester interchange reaction. These data are shown in Table II and together with the aforementioned data, indicate that this pair is immiscible, although not fully so, and that transreaction has to take place in the PAr-rich phase. These results are in good agreement with those obtained by Robeson⁴² and show that by an adequate control of the processing temperature it is possible to produce blends with different transesterification levels; this can even be possible at high temperatures in a normal molding process.^{42, 43}

To analyze the influence of interchange reactions between both polymers on the mechanical behavior of the blend, the tensile properties have been studied before and after transesterification. As can be observed in Figure 4, Young's modulus of the different compositions is close to a tie line connecting the pure

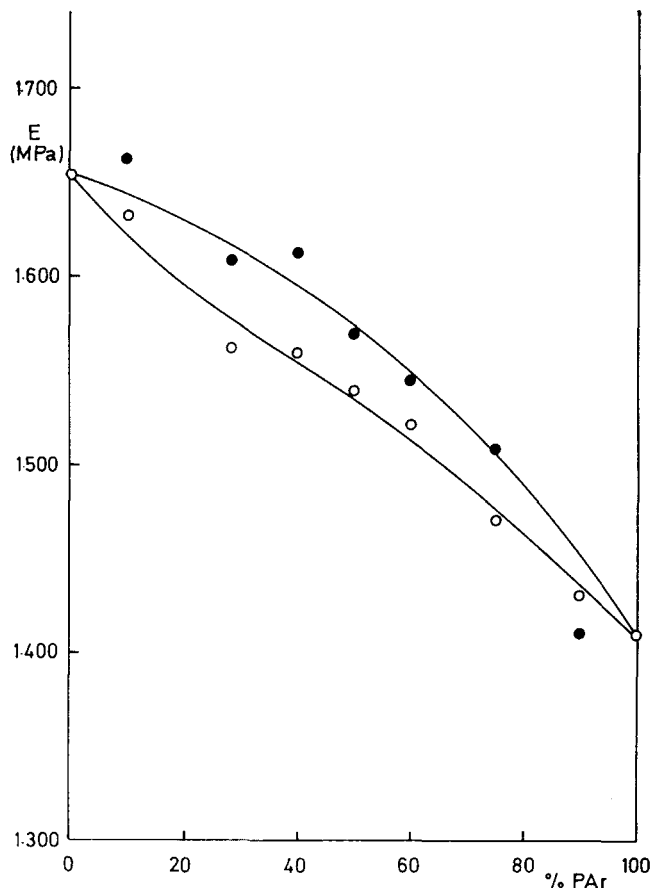


Fig. 4. Young's modulus of before (○) and after (●) transesterification mixtures against blend composition.

component values, which is indicative of some miscibility or, at least, of the fact that there exists a good adhesion between both polymers. Moreover, it is observed that the modulus value is above this line at PAr-rich compositions. This could be explained if we consider that miscibility increases when the PAr content is higher, but as has been shown above, it can also be attributed to the formation of some quantity of copolymer at these PAr-rich compositions because the processing method used allows for the obtaining of higher melt temperatures at PAr-rich contents. The copolymer obtained can act as a compatibilizing agent between two phases thus improving the mechanical properties of the blend.

If the values corresponding to post-transesterification blends are analyzed, the generalized synergistic effect of transesterification on the modulus can be observed. This synergism can be quantified by the equation that has been applied to miscible blends⁴⁴⁻⁴⁷

$$E = w_1 E_1 + w_2 E_2 + \beta_{12} w_1 w_2$$

where E is the blend modulus, E_1 and E_2 the pure component modulus

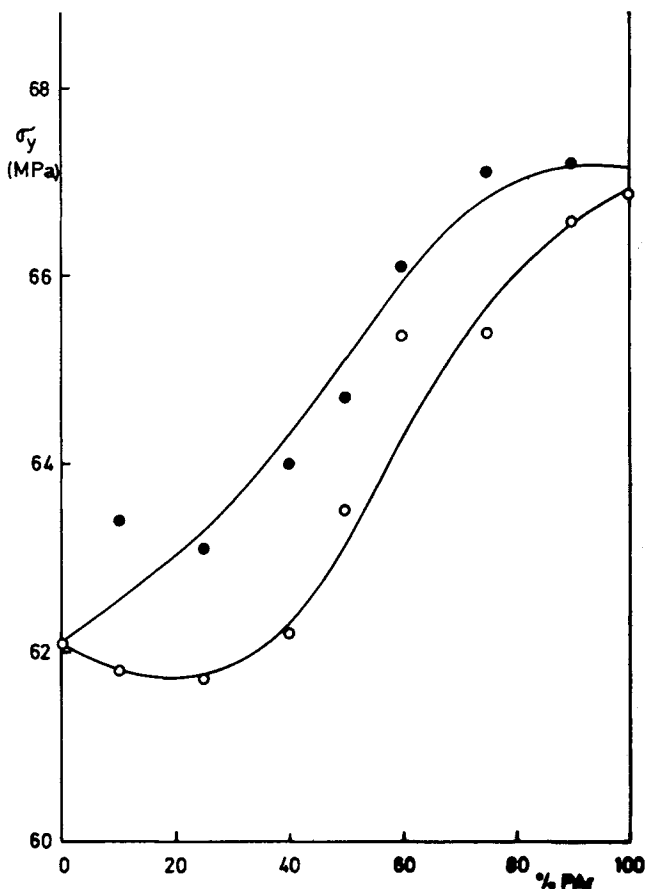


Fig. 5. Yield strength vs. blend composition: (○) blend; (●) copolymer.

values, w_1 and w_2 the weight fractions, and β_{12} an empirical interaction term, which can be calculated as

$$\beta_{12} = 4E_{12} - 2E_1 - 2E_2$$

As has been suggested in some papers,^{25,46,47} high-strain mechanical properties, yield strength, and mainly stress and strain at break, were found to be strongly dependent on the blend miscibility, and are more indicative of the interfacial adhesion between components.

This is the reason for showing in Figure 5 the yield stress values of the blend before transesterification against the blend composition. The results are quite different with respect to the modulus data and show the greater effect of the immiscibility on this property. This is supported by the clear minimum that is observed at about 25% PA_r and explains the lack of transparency in the low PA_r content blends. On the other hand, at high PA_r compositions, the compatibilizing effect of the copolymer formed in the first step of the reaction appears to be strong enough to carry the values of the yield stress close to the linearity.

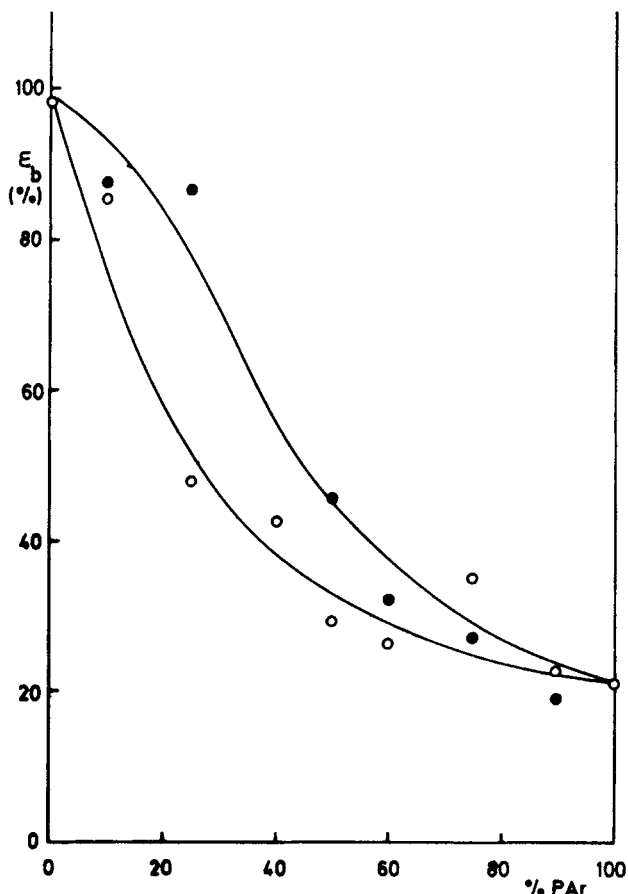


Fig. 6. Deformation at break vs. blend composition: (○) blends; (●) copolymer.

As shown in Figure 5, the yield stress results corresponding to the reacted copolymer appear practically on the tie line connecting the pure polymer values, and they are also higher than those of the pretransesterification blends at all compositions. This is due to the loss of free volume as a consequence of the new copolymer structure developed in melt mixing.

The tensile properties that are more susceptible to miscibility level in the blend are stress and strain at break. To observe the variation of deformation at break, it has been drawn vs. blend composition in Figure 6. From these data it is clear that these polymers are immiscible. However, as happens with other tensile properties, at PAr-rich compositions, blends, and transesterified-mixture values are quite similar. This confirms the above considerations about a fast first step of transesterification that at these compositions is relatively important with respect to the overall blend; this copolymer that results from the transesterification will considerably improve the adhesion between the two immiscible phases even up to a point where miscibility is attained. The strain at break values of the reacted blends are closer to the values corresponding to the linear rule of mixtures and again show that the copolymer developed has a structure that differs totally from that of the nontrans-

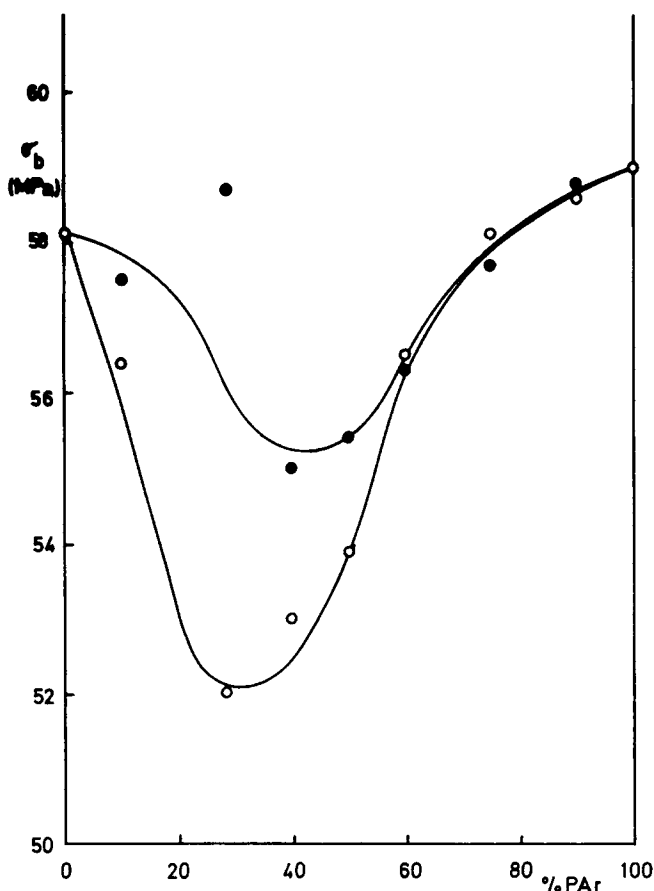


Fig. 7. Strength at break against blend content: (○) blend; (●) copolymer.

esterified blend. The interchange reaction does not produce crosslinking between both polymers since, if they were to crosslink, the copolymers formed would have a very low deformability, and, as has been shown, this does not occur.

The data of stress at break shown in Figure 7 corroborate the immiscibility of this pair because of the existence of a minimum in the stress at about 25% PAr. The transesterified blend values are, as has already been mentioned above, higher than those of the mixtures before the interchange reaction. As for the behavior of the other tensile properties, at high PAr contents, the mixture values corresponding to maximum and steadied torque are quite similar. This confirms the supposition previously made about the course of transesterification.

CONCLUSIONS

1. The PAr/PC pair is immiscible at all blend compositions as observed in solvent-cast blends.

2. In this blend interchange reactions can be produced under normal processing conditions, obtaining transesterified copolymers which show one single T_g at all mixture contents.

3. The exchange reaction can be controlled by adequate choice of the processing temperature, thus allowing for the obtaining of a copolymer at different residence times.

4. All the mechanical properties of the copolymers that have been studied are higher than those of the corresponding physical blends, and a major increase at PC-rich compositions is observed.

5. Transesterification in these blends seems to have taken place in two steps: The first one develops in the PAR-rich phase, and the second one develops between both immiscible phases.

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