

Interfacing Polypropylene/Poly(BPA carbonate) Blend Phases with Graft Copolymers: Thermal and Dynamic Mechanical Analyses*

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SYNOPSIS

This paper presents data on the thermal, dynamic, and mechanical properties of polypropylene (PP) blends with bisphenol-A-polycarbonate (PC), to which a series of graft and block copolymers in a small quantity was added. The effect of the minor component on the crystallization and relaxation behavior of PP in the blends has been investigated and correlated with the mechanical properties obtained. The results demonstrate that the graft copolymeric additives to the blends can reduce the degree of undercooling ($T_m^0C - T_c^0C$) of the PP phase. However, the block copolymers used, substituting for the graft copolymers, showed no such function. Dynamic mechanical thermal analysis (DMTA) indicates that, with the addition of several different copolymers as minor additives to the blends, two loss peaks representing the glassy transitions (T_g) of the individual components (PP and PC) were retained, with little tendency of approaching toward each other, suggesting no obvious improvement in compatibility of the PP phase and the PC phase in the blends. Nevertheless, the inclusion of different copolymers in the PP/PC blends, in spite of the small quantity used (4%), can lead to a significant mechanical property difference of the blends. This difference could be reasonably explained from the data obtained in dynamic mechanical characterization of the different graft copolymers in PP blends.

INTRODUCTION

Because of its low cost and versatile properties, polypropylene (PP) is a widely used thermoplastic. Ethylene-propylene-diene terpolymer (EPDM)-modified PP blends with high impact strength, which were developed commercially, have widened PP's applicability considerably. The impact/stiffness balance and the heat-distortion temperature of general grades of PP products are far inferior to those of most engineering plastics. These deficiencies have limited its application in many fields. Therefore, research and development of the engi-

neered PP materials is attracting more and more interest.

Polypropylene blends with high-performance engineering resins could be expected to improve the deficiencies inherent in PP and to reach an unique balance of the performance/cost ratio and further upgrade the use of the PP series. Up to now, although a number of PP-based blends with engineering resins [such as PC, poly(ethylene terephthalate) (PET) and polyphenylene ether (PPO)] have been reported on their mechanical properties,¹⁻⁵ permeability,⁶ rheological,^{7,8} and dielectric behavior,⁹ the results obtained indicated that PP blends had poor mechanical behavior, severe delamination, and very weak weld line strength in their extrusion or injection-molded parts. Admittedly, a lack of adhesion at the interface was considered mainly responsible for the inferior performance. A possible solution to this problem could be addition of a selected copoly-

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meric additive as a minor third component that can promote adhesion at interphases of the blends. However, the presence of the third components, although in only a small quantity, might also bring about some other significant changes such as in crystalline behavior, morphology, and dynamic mechanical and rheological properties. The blend properties could be influenced through a more complicated pattern in different ways, depending on several factors. These effects are not readily predictable.

This paper presents data on the thermal, dynamic, and mechanical properties of PP blends with PC, in which a series of graft and block copolymers (PP-*g*-styrene [PP-*g*-St], PP-*g*-St/acrylonitrile [PP-*g*-St/AN], PP-*g*-styrene/butyl methacrylate [PP-*g*-St/BuMA], PP-*b*-butyl methacrylate [PP-*b*-BuMA], and styrene-*b*-ethylene-*co*-butylene-*b*-styrene [SEBS]) in small quantities (4% wt based on the PP/PC blends) were added. The effects of each of the minor components on the crystallization and relaxation behavior of PP in the blends have been examined and correlated with the observed rheological and mechanical properties.

EXPERIMENTAL

Blend Sample Preparation

The polypropylene used in this study was Moplen D 50S produced by Himont, Italy. The bisphenol-A-polycarbonate used was Sinvet 251 manufactured by Enichem, Italy. The copolymeric additives involved in the work were SEBS triblock copolymer (Kraton G 1652 by Shell) and some PP-based graft copolymers synthesized on a laboratory scale. The information on them is shown in Table I.

Polymer blends were prepared by melt mixing. The mixing conditions ($T^{\circ}\text{C}$, shear rate $\dot{\gamma}$) were

defined according to the viscosity matching principle, based on the rheological data [shown in Fig. 1(a) and (b)] by means of the capillary rheometer (Instron type TTCM/AQ 723). Before mixing, all components were dried in a vacuum oven at 80°C (for PP and copolymers) and at 130°C (for PC) for 8 h. In order to ascertain uniform dispersion of the minor component in the PP/PC ternary blends, PP/copolymer (PP-Cop) masterbatches were prepared first in a Brabender mixer (W50) at 220°C and 20 rpm with a mixing time of 10 min. The blends of desired composition from PP, PC, and PP-Cop masterbatches were compounded at 270°C and 20 rpm under N_2 protection in a 25/1 L/D screw extruder (Brabender PLD 651). Then, the blends obtained in the extruder, which had been palletized and dried, were compression molded at 270°C into the plates or sheets from which the standard test specimens were obtained.

Measurements

The melting and crystallization behavior of the pure components and blends were investigated using a DuPont Model 910 differential scanning calorimeter (DSC). DSC measurements were carried out in N_2 by heating the sample isothermally at 200°C for 5 min (to impart identical thermal history to all the pellets), followed by cooling at $20^{\circ}\text{C}/\text{min}$ to determine the crystallization temperature (T_c) and then heating from room temperature to 200°C at $10^{\circ}\text{C}/\text{min}$ to determine the melting thermograms.

The dynamic mechanical properties were determined using a Polymer Labs PL-DMTA. Samples ($40 \times 10 \times 1.5$ mm approx.) were cut from the compression-molded sheets. Measurements were made at an operating frequency of 1 Hz and a heating rate of $2^{\circ}\text{C}/\text{min}$ with the samples in a nitrogen atmosphere.

Table I Copolymeric Additives Involved in PP Blends with PC

Code	Copolymer Type	Composition (wt %)		
Cop-1	PP- <i>g</i> -St	82% PP	8% St grafted	10% St ungrafted
Cop-2	PP- <i>g</i> -St/AN	90% PP	2.5% St/AN ^a grafted	7.5% St/AN ungrafted
Cop-3	PP- <i>g</i> -St/ <i>n</i> BuMA	86% PP	7% St/ <i>n</i> BuMA ^b grafted	7% St/ <i>n</i> BuMA ungrafted
Cop-4	PP- <i>b</i> - <i>n</i> BuMA ^c	91% PP	9% <i>n</i> BuMA	
Cop-5	SEBS (Kraton)	30% PS	70% EB copolymer ^d	

^a 20% AN in St/AN (by wt).

^b 70% *n*BuMA in St/*n*BuMA (by wt).

^c Wax, MFR: 50 g/10 min (ASTM D 1238L).

^d 52% E in EB (by wt).

The mechanical properties, including stress-strain properties, notched Izod impact strength, heat-distortion temperature (HDT), and flexural modulus were measured according to ASTM D638, D256, D648 and D790, respectively. The melt flow rate (MFR) was measured using ASTM D1238L (230°C, 2.16 kg) and D12380 (300°C, 1.2 kg) conditions.

RESULTS AND DISCUSSION

Processing and Rheology

Since the processing conditions and the rheological characteristics of the components are important factors that affect the blend morphology and thereby the properties of the end products, the rheological behavior of the PP and the PC polymer components were observed first by means of a capillary rheometer (Instron TTCM/AQ723).

The rheological curves, shown in Figure 1(a), illustrate that there is a large disparity in the flow behavior between PP (Moplen D) and PC (Sinvet 251). The PC apparent melt viscosity (η) is much more temperature dependent, whereas (η) for PP is more sensitive to variation of shear rate. Therefore, an optimal process condition to obtain uniform dispersion could be chosen at which the viscosities of the PP and the PC may match.¹⁰ Figure 1(b) clearly shows that, in the appropriate temperature and shear-rate range, one can find a large viscosity overlapping zone of PP and PC in accord with the demands of both stable blending technology and low equipment power requirements.

In Figure 2, the melt flow rate (MFR) values of PP blends and the blends with a small quantity of Cop-3 (PP-*g*-St/BuMA) additive, measured under "L" and "O" testing conditions, are reported as a function of the PC content. The results show that a maximum on the MFR-composition curve exists at about equal composition (50/50) at the "L" temperature condition; however, at a higher testing temperature (300°C at "O" condition), the location of the maximum on the curve was shifted to higher PC content at about 75% PC because of the PC's larger temperature dependence according to the results in Figure 1. The rheological data (shown in Fig. 3), obtained from measuring stable torque values of the samples during melt blending by a Brabender Plasticorder, also coincide with the above MFR results: a minimum on the torque-composition curve existing at about equal blend composition.

In general, as reported by some authors,^{11,12} for

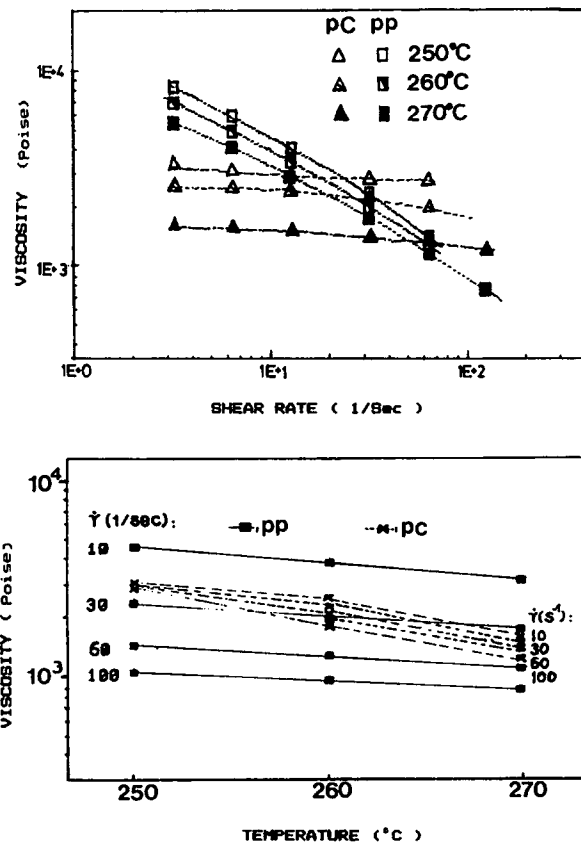


Figure 1 (a) Melt viscosity versus shear rate at different temperatures for PP (Moplen D) and PC (Sinvet 251). (b) Viscosity of PP and PC as a function of temperature in the shear rate range (40–100 s⁻¹).

incompatible blend systems with varying blending ratios, the melt viscosity of blends goes through a minimum, based on the idea of a reciprocal lubricating effect of polymeric components. From this viewpoint, Cop-3 used as a minor third component, which would be expected to stay at the interface of the PP phase and the PC phase, did not alleviate this reciprocal lubricating effect, but on the contrary, adding it led to a lower viscosity minimum value at the same composition (see Figs. 2 and 3). In Figure 4, zone-II may be considered as a contribution to viscosity reduction resulting from a synergistic effect of Cop-3 in the PP/PC ternary blends, whereas zone-I may reflect a contribution to decreasing viscosity due to PC added in the PP blends.

The effect of several different copolymers (Cop-1 to Cop-5) as third components respectively, on the flow behavior of the PP/PC blends is shown in Figure 5. The results indicate that the influence on the flow properties of the blends are very different, depending on the sort of copolymeric additives used.

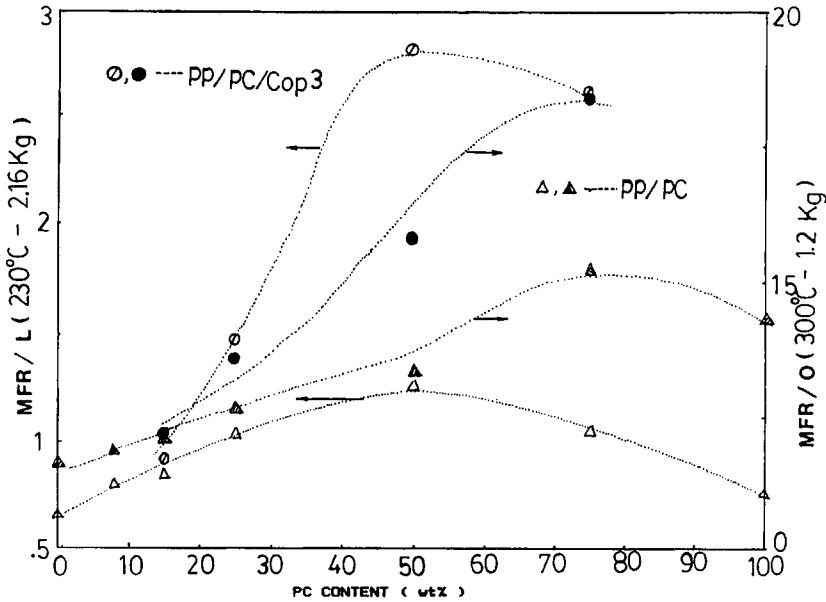


Figure 2 Melt-flow rate (g/10 min) versus PC content (wt %) for the PP/PC blends with and without Cop-3 (4%).

Cop-3 and Cop-4 (both with BuMA units), especially Cop-4 with high fluidity, could function effectively as viscous diluents for the PP/PC blends at low content (lower than 4% wt based on the blends). However, under the same condition, Cop-2 with St/AN produced a much less viscous reduction effect by comparison, which seems to be advantageous to the promotion of interface adhesion in incompatible binary blending systems from the viewpoint of the reciprocal lubricating effect.¹¹

Crystalline Property

The DSC analysis data on PP (Moplen D), its blends with PC, and the blends with several copolymeric additives (Cop-1 to Cop-5), respectively, are shown in Tables II and III. The results in Table II indicate that PP crystallization was not affected by the presence of the PC phase in both binary and ternary blends. As a consequence of that, the melting temperatures (T_m) as well as the crystallization

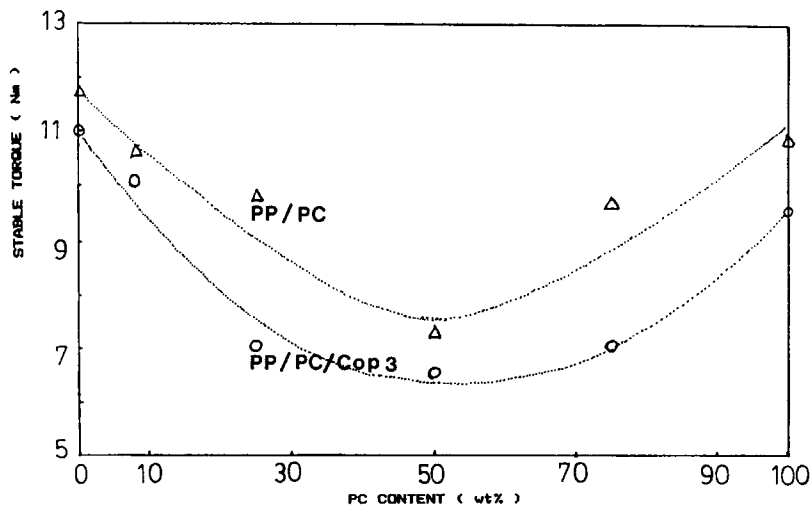


Figure 3 Effect of PC content on viscosity of the PP/PC blends with and without Cop-3 (4%).

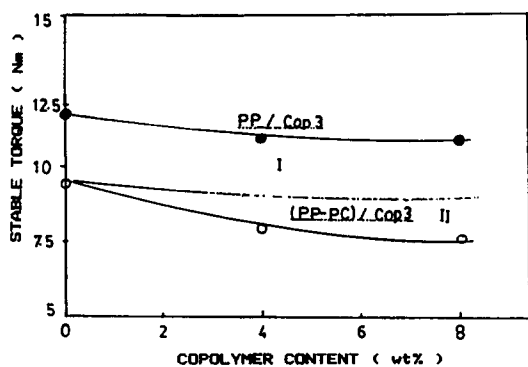


Figure 4 Viscosity as a function of Cop-3 content in PP and the PP/PC (75/25) blends.

temperatures (T_c) of the PP phase in the blends were unchanged with increasing PC content. However, the presence of Cop-3 in the blends could affect the PP crystallization in a certain way: The T_c of the PP phase was increased significantly, while its T_m was not changed basically, resulting in a lower degree of supercooling and a higher crystallization rate. Moreover, the T_c -enhancement of the PP was not sensitive to the variation of Cop-3/PP ratio in the ternary blends with changing PC content, indicating that Cop-3, effective already at low concentration, acted as a fair nucleating agent to increase the PP crystallization temperature.

Data on the heat of fusion of the binary and ternary blends are summarized in Figure 6, where the normalized quantity $[1 - H_f(\text{blend})/H_f(\text{PP})]$, tak-

ing into account the concentration effect, was plotted against PC content (% wt). It can be inferred from this figure that the incorporation of a certain amount of Cop-3 (PP-*g*-St/BuMA), despite the T_c increase of the PP phase and the lower degree of undercooling therefrom, resulted in no appreciable change in the heat of fusion or the degree of crystallinity other than a trivial volume effect because all data were near the diagonal line representing the situation where the PC (or PC/Cop-3) in the PP blends has no effect on the heat of fusion of PP.

In Table III, the effects of several different copolymers added in the PP/PC blends were compared on the crystallization of the PP matrix. Similar to the effect of Cop-3 (PP-*g*-St/BuMA), two other PP graft copolymers (Cop-1: PP-*g*-St, Cop-2: PP-*g*-St/AN) can function as fair nucleating agents in the same way to obviously increase T_c of the PP blends, whereas both Cop-4 (PP-*b*-BuMA) and Cop-5 (SEBS) have not as appreciable a function in increasing T_c of PP under the same conditions. Obviously, the above phenomenon could be related to the molecular structure features of copolymeric additives used.

It is known^{13,14} that in crystallization behavior there exist obvious disparities between PP homopolymer and PP graft copolymer, not only in the temperature and rate of crystallization but also in their crystalline morphologies, which could be attributed to the adsorption and disturbance on the crystal surface by graft chains of PP. These probably

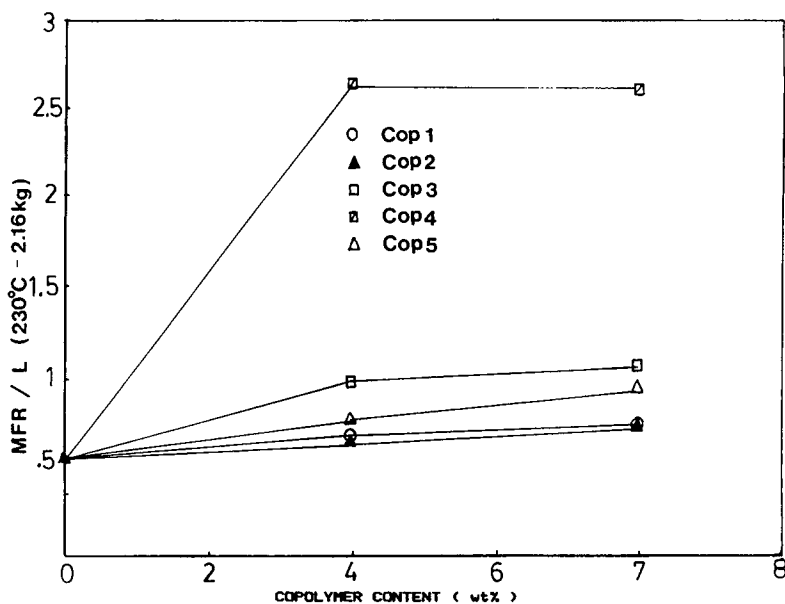


Figure 5 Effect of copolymer type on MFR of the PP/PC (75/25) blends.

Table II DSC Melting and Crystallization Results of PP, PP/PC, and PP/PC/Cop-3 Blends

Samples and Composition	Melting Peak (°C)	Heat of Fusion (J/g)	Crystallization Peak (°C)	Degree of Undercooling (T_m °C - T_c °C)
PP polymer (Moplen D)	159.0	81.0	106.0	53.0
PP/PC (92/8)	158.8	75.0	106.0	52.5
PP/PC (75/25)	159.0	67.0	107.0	52.0
PP/PC (50/50)	158.5	39.0	107.5	51.0
PP/PC/Cop-3 (88.3/7.7/4)	159.5	74.0	112.0	47.5
PP/PC/Cop-3 (72/24/4)	159.5	63.0	112.0	48.0
PP/PC/Cop-3 (48/48/4)	157.5	40.5	110.5	47.0

diverted the PP normal spherulitic three-dimensional developing characteristics into a rodlike two-dimensional crystalline morphology.¹³ In the PP/PP-*g*-copolymer/PC blends, the quite strong adhesion between the two (PP homopolymer and graft copolymer) crystalline phases can stem from the highly compatible amorphous phases. The PP-graft copolymers may act as nucleating agents to cocrystallize with the PP phase like the nucleation mechanism of the PP-graft-acrylic acid copolymeric additive (Polybond, BP Chemicals) to the PP blends,¹⁵ if the crystallization temperatures of the PP and the PP graft copolymer are not far apart. Also, the graft copolymers may directly play the role of impurity to provide the primary nuclei at the center of the

spherulite incorporated into the crystal or absorbed onto the crystal. Either way, it suffices to have a small number of nuclei affecting the PP crystallization behavior. Direct evidence has not been gathered to confirm the nucleation mechanism discussed because of the need of further investigation into the graft copolymers used for their crystallization behavior in detail. However, the PP-*b*-BuMA (Cop-4) used in this study has high fluidity, is a very good diluent for the PP blend (see the rheological data), and behaves like a wax with very low mechanical strength at room temperature. On cooling of the PP/PC/Cop-4 blend, at the temperature at which PP crystallizes, the Cop-4 is still a liquid, so it cannot serve as a nucleating agent in normal considera-

Table III Effect of Additive Type on Melting and Crystallization of PP in the Blends

Samples and Composition	Melting Peak (°C)	Heat of Fusion (J/g)	Crystallization Peak (°C)	Degree of Undercooling (T_m °C - T_c °C)
PP polymer (Moplen D)	159.0	81.0	106.0	53.0
PP/PC (75/25)	159.0	67.0	107.0	52.0
PP/PC/Cop-1 (72/24/4)	158.5	60.0	114.5	44.0
PP/PC/Cop-2 (72/24/4)	160.0	70.0	115.5	43.5
PP/PC/Cop-3 (72/24/4)	159.5	63.0	112.0	48.0
PP/PC/Cop-4 (72/24/4)	159.0	70.5	109.0	50.0
PP/PC/Cop-5 (72/24/4)	159.0	57.0	107.5	52.0

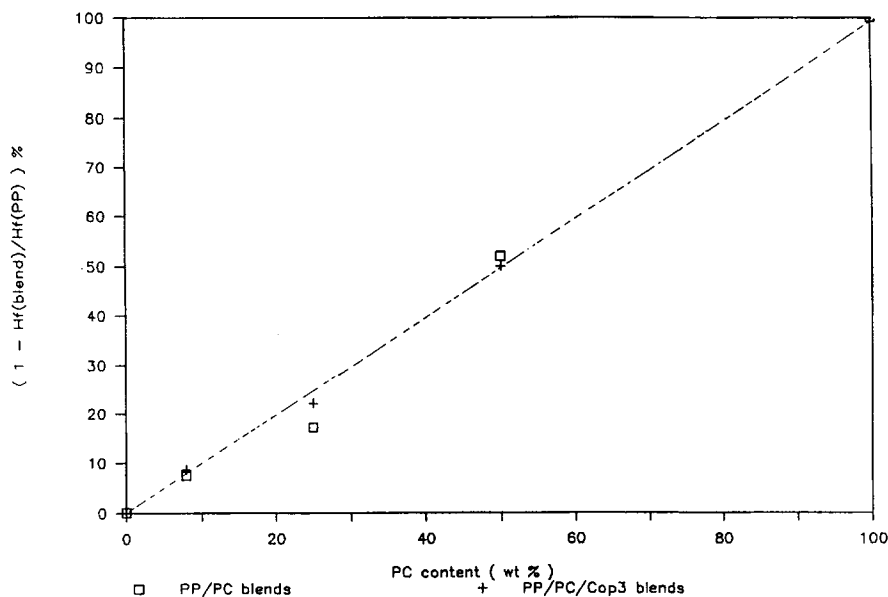


Figure 6 Data on heat of fusion of the PP phases in the PP/PC/Cop-3 blends.

tion.¹⁶ The Cop-5 (SEBS elastomer) has no possibility of showing nucleation by cocrystallization with PP due to its molecular structure. Although a number of impact modifiers (like EPR, EPDM, and SBR, etc.) in the PP matrix do function as a nucleating agent through playing the role of an impurity to increase T_c of PP and lower the degree of undercooling of PP crystallization,^{17,18} SEBS (Kraton 1652) as a minor third component in the PP/PC ternary blends could be expected to partially stay at the PP-PC interface and/or enter into the PC phase and thereby reduce its nucleating efficiency, if any, to influence PP phase crystallization. The data obtained by A. K. Gupta's research on the crystallization of PP in PP/SEBS blends¹⁹ also indicated that SEBS (Kraton 1652) inclusion in the concentration range up to 20% by wt did not give rise to an obviously changed T_c of the PP matrix crystallization.

Dynamic Mechanical Properties

The dynamic mechanical data for PP, PC, and their binary blend, viz. the storage modulus E' and loss tangent $\tan \delta$ as a function of temperature, are shown in Figure 7.

The data indicate that PP has a higher storage modulus E' than does PC in the low-temperature zone until the onset of the PP damping peak at about 0°C and, afterward, drops down gradually by about an order of magnitude with increasing temperature up to 100°C. The $\tan \delta$ curve of the PC homopolymer

shows two mechanical transitions: the γ -transition at about -95°C and the β -transition at 152°C. The β -transition of PC corresponds to its glass transition, while the γ -transition is associated with the in-chain

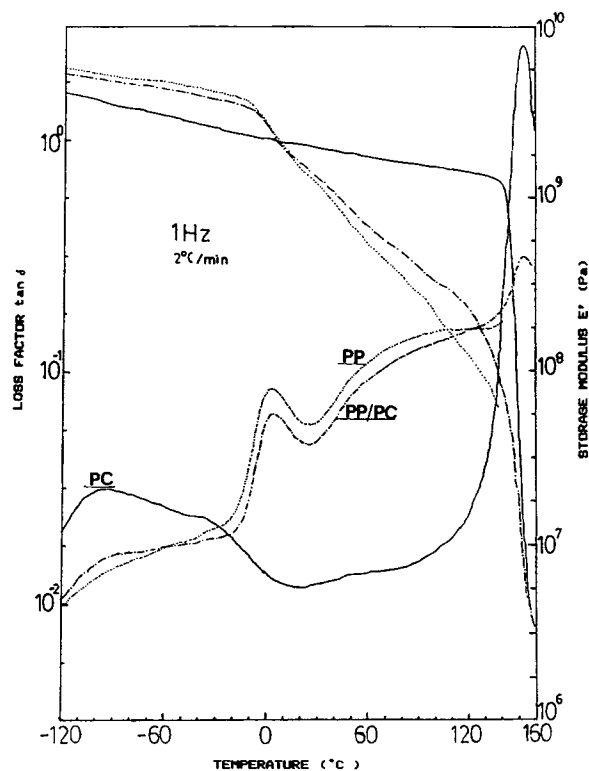


Figure 7 Dynamic mechanical properties of PP, PC, and the PP/PC (75/25) blend.

motion of the carbonate group of PC. On the PP $\tan \delta$ curve, the β -transition of PP that represents its T_g appears at about 4°C.

It can be seen clearly from Figure 7 that the PP/PC (75/25) blend exhibited two damping peaks at 4 and 152°C, corresponding to the glass transition temperatures of PP and PC homopolymers, respectively, while the γ -transition peak for PC became much less distinct. These data indicate the immiscibility of the components and phase separation during melt blending to give phases of pure PP and PC at this composition.

Figure 8 illustrates the influence of different copolymer additives on the dynamic mechanical properties of the PP/PC blends. The relaxation behaviors of all ternary blends are similar to that of the binary blend without copolymer as a minor third component, except for the PP/PC/Cop-5 blend, which showed a weak damping peak (centered at around -42°C), attributable to the motion of the EB random segments in Cop-5 (SEBS). The presence of all PP-based copolymers involved did not result in any significant influence on the dynamic

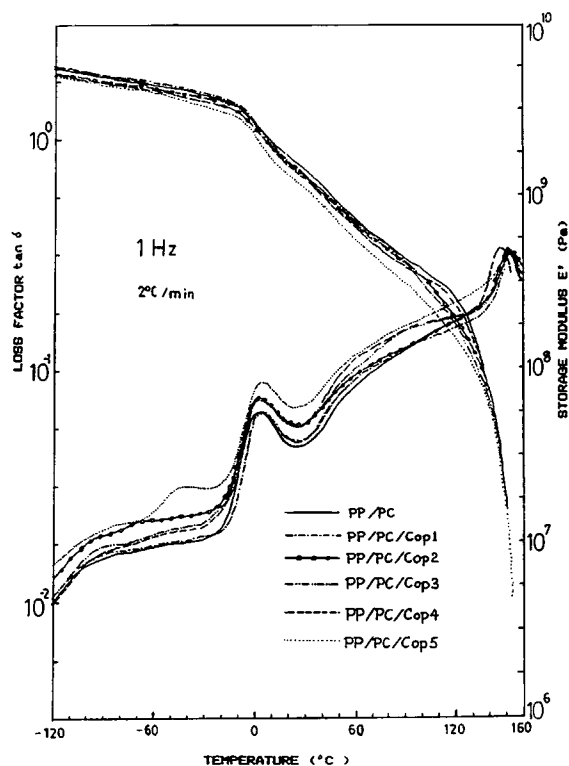


Figure 8 Dynamic mechanical properties of the PP/PC (75/25) binary and the PP/PC/copolymer (73/24/4) ternary blends (Cop-1: PP-*g*-S; Cop-2: PP-*g*-S/AN; Cop-3: PP-*g*-St/BuMA; Cop-4: PP-*b*-BuMA; Cop-5: SEBS).

mechanical properties of the PP/PC blends at least at the level of the amount added. The two loss peaks representing glass transitions of the two main components are still present in the ternary blends, with little tendency to approach each other, commonly meaning that the copolymers added could not bring obvious improvement in compatibility between the PP phase and the PC phase in the blends.¹

The PP copolymers used in the study were selected by considering their possible action and difference as compatibilizers for the PP/PC blend, based on the solubility parameters of the grafted chains (St, St/BuMA) being close to that of PC, or on the grafted chains having the same molecular structure as the copolymers (like St/AN) that have good adhesion with PC according to reports,^{20,21} while the PP backbone should be miscible in the PP matrix. However, the results obtained from dynamic mechanical analysis did not suggest such a function of the copolymers as adhesion modifiers for the PP/PC system, in spite of the molecular structures that suggested their ability to do so. The question arises whether these PP copolymers, as minor components, can exert a significantly different effect on the mechanical properties of the PP/PC blends since their existence, in spite of a small quantity (only 4% wt), brought about appreciable effects on the rheological and crystallization properties of the PP-based blends with PC according to the above sections.

Before going into the next section (mechanical properties), further dynamic mechanical analysis is made on the PP-*g*-copolymers (Cop-1, Cop-2, and Cop-3), which were blended with PP homopolymer, respectively, at an equal ratio, hoping to understand further the relaxation features of PP graft copolymers and the interaction between graft copolymers and PP homopolymer. The results, shown in Figure 9, indicate a common feature of $\tan \delta$ versus T curves of the three PP/PP-*g*-copolymer blends, that is, the location of β -relaxation peaks (or T_g) of PP segments shifted from 4°C to higher temperature of 6–7°C, which may have resulted from the action of the PP backbone of the copolymers on the PP homopolymer.

The existence of the other loss peaks in Figure 9 should be attributed to the contribution of other branched chains of PP graft copolymers in the blends. On the curve of PP/Cop-1, the damping peak at about 100°C corresponds to the β -transition of polystyrene (PS) segments in PP-*g*-styrene. In Cop-2, inserted into the PS branched chain was about 20% by wt acrylonitrile (AN) and the peak at 100°C for PS segments was shifted to higher temperature at about 130°C, as shown on the $\tan \delta$ ver-

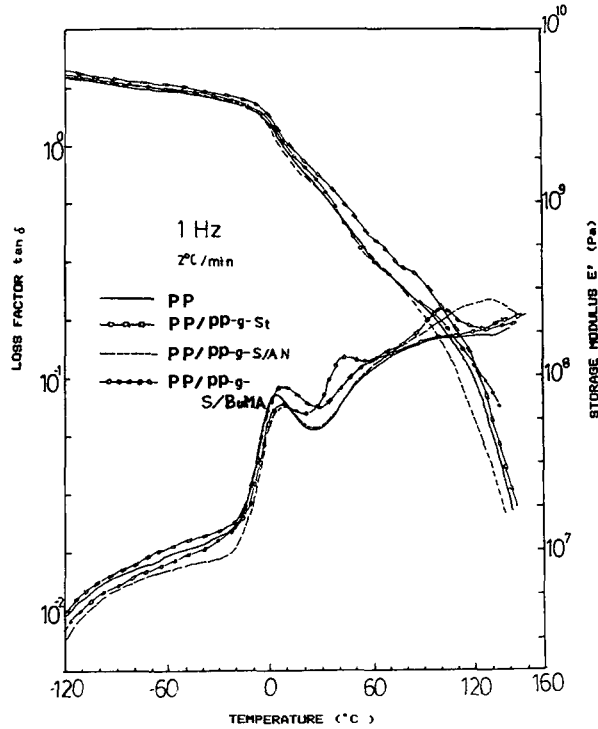


Figure 9 Temperature dependence of $\tan \delta$ and E' for PP and the PP blends (50/50) with PP-*g*-copolymers (Cop-1: PP-*g*-St; Cop-2: PP-*g*-St/AN; Cop-3: PP-*g*-St/BuMA).

the $\tan \delta$ versus T curve of the PP/Cop-2 sample, in which the broad peak in the 120 to 140°C range has probably arisen from segmental motion of St/AN random branched chains with higher polarity than that of

the PS segments. Introducing *n*-butyl methacrylate (*n*BuMA) units (about 70% by wt) into the PS branch chain rather than the AN unit can greatly increase the flexibility of the PS branched chains. Thus, on the $\tan \delta$ versus T curve of the PP/Cop-3 sample, a damping peak appeared at around 43°C with the β -transition peak of the (*S*) segments disappearing. The measured value for St/*n*BuMA branched chains is basically consistent with the calculated value according to the additive formula for random copolymerization at the same ratio.

From the above analysis, it can be shown that the slight structural adjustments (styrene, styrene/acrylonitrile, and styrene/butyl methacrylate) of branch chains grafted onto the PP backbones can give rise to obvious variation in the relaxation behavior of PP materials. This variation, in combination with changes in the rheological and sometimes crystalline behavior of the PP blends due to the presence of copolymeric additives, should cause obvious effects on the mechanical properties of the PP-based blends with PC.

Mechanical Properties

PC is an engineering plastic with outstanding mechanical strength and temperature resistance. The heat-distortion temperature (HDT) of PC at 1820 kPa is around 140°C. Therefore, added PC in the PP resin can improve the heat resistance of the materials, thus upgrading the performance such as the endurance of the material at elevated temperatures.

Figure 10 gives the HDT (at 1820 kPa) of PP,

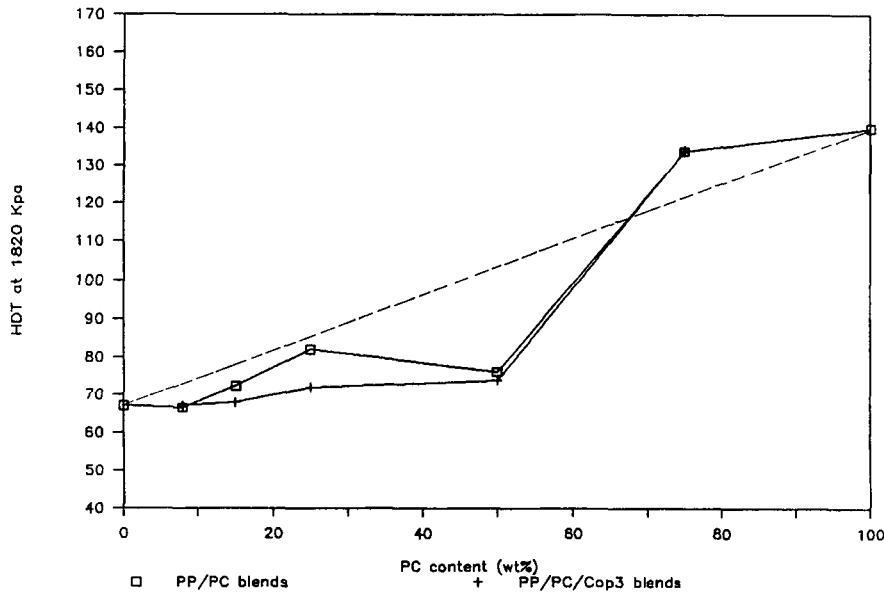


Figure 10 Heat-distortion temperature of PP, PC, and PP/PC/Cop-3.

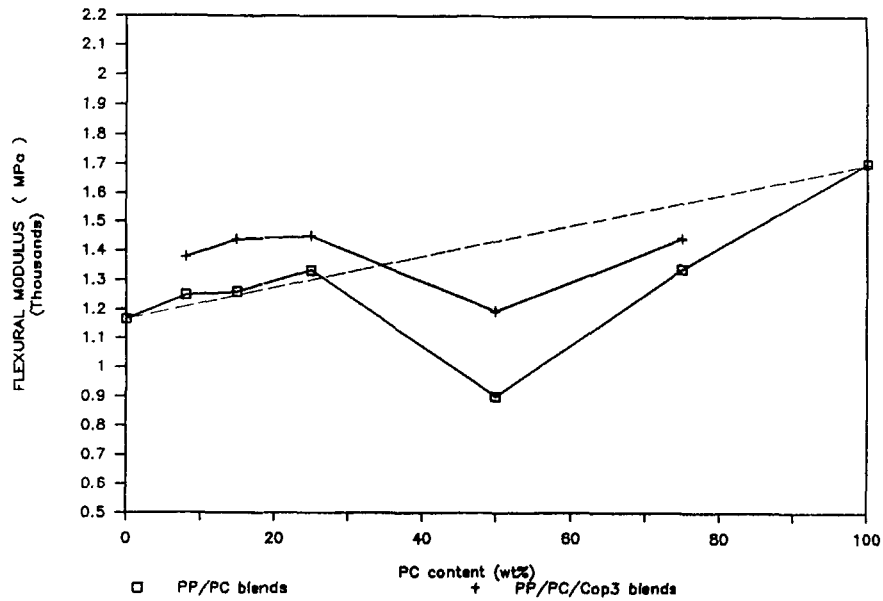


Figure 11 Flexural modulus of elasticity of PP, PC, and PP/PC and PP/PC/Cop-3 blends.

PC, the PP/PC blends, and the blends with Cop-3 (4% wt). The data indicate that the HDT increased with increasing PC content, as expected. However, if we use a rule of mixtures to judge the blending effect, as follows:

$$P = P_a\phi_a + P_b(1 - \phi_a) \quad (1)$$

where P_a , P_b , and P are the measured properties of a and b components and blend, respectively, and ϕ_a

is the weight fraction of the a -component in the blend, then the HDT properties of the blends (both binary and ternary) show an obvious negative deviation from the values estimated from eq. (1) when the concentration of two components in the blends are comparable. The presence of Cop-3 in the blends further reduced the HDT of the PP-based blends in the lower range of PC content.

The flexural moduli (FM) of the above samples are given in Figure 11. In the low range of PC up to

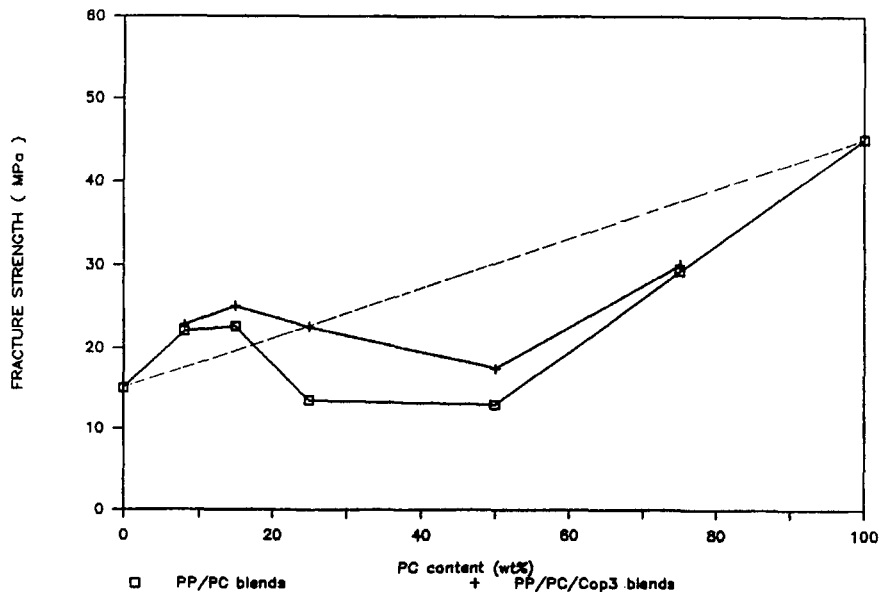


Figure 12 Tensile strength (MPa) at break of PP, PC, PP/PC, and PP/PC/Cop-3.

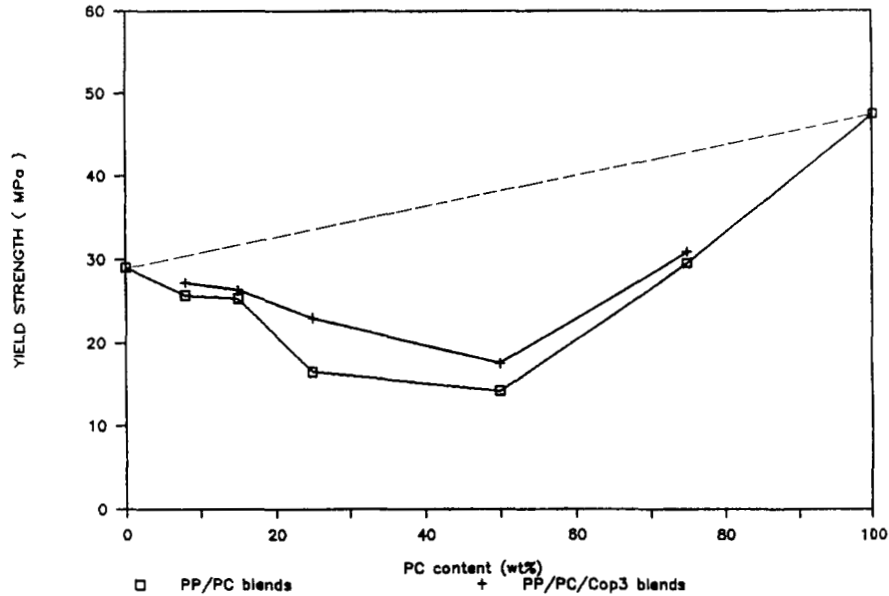


Figure 13 Tensile strength (MPa) at yield of PP, PC, PP/PC, and PP/PC/Cop-3.

25%, the FM value is increased, showing a weakly positive blending effect; thus, it is slightly higher than the value estimated from eq. (1), and then it decreases quickly until at about 50/50 blending ratio. Afterward, the FM value goes up rapidly with decreasing PP content. Obviously, this property change may arise from the blend morphological phase reversion at about equal composition, representing the poorest balance of properties in the incompatible blend system.²¹ The curves of fracture strength versus composition of PP/PC or PP/PC/Cop-3 blends also show a similar trend basically (see

data in Fig. 12). Figure 13 gives the tensile yield values of the samples as a function of blend ratio. Although a strength minimum value also took place at about the phase transition composition, a negative blending effect in the tensile yield was found in the PP/PC blends in the whole range of composition. Clearly, it was due to the weak interfacial bonding between PP and PC that the high-strength property of PC did not offer any advantage of improving the strength of the blends, especially in the PP-PC phase transition composition range. Addition of Cop-3 (PP-*g*-St/BuMA) in the PP/PC blends did

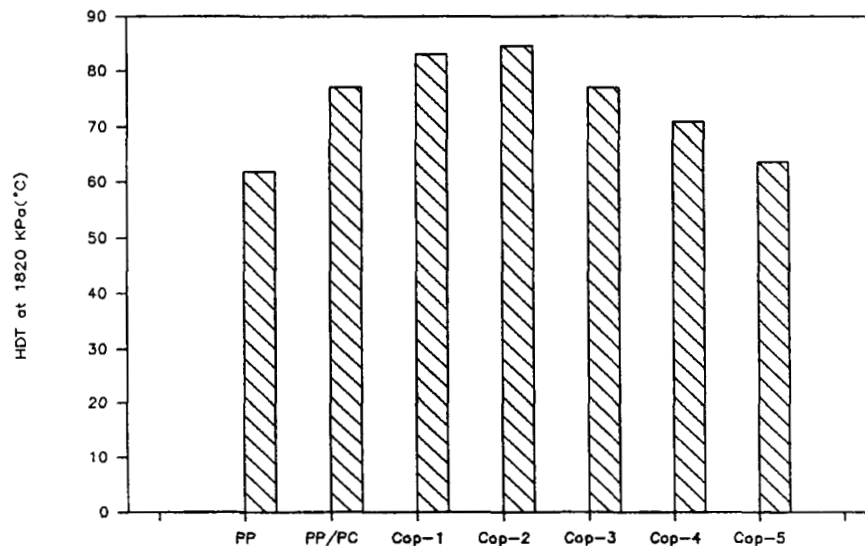


Figure 14 Effect of copolymers added (4%) on HDT of the PP/PC (75/25) blends.

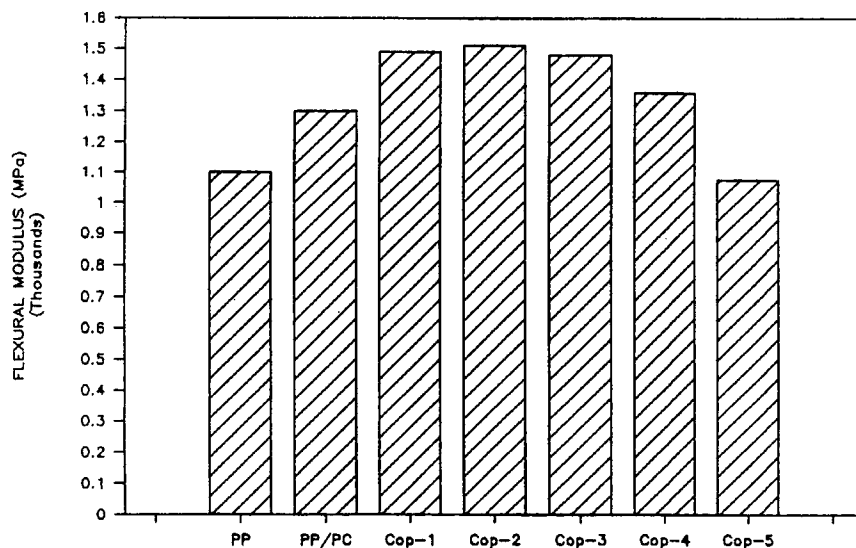


Figure 15 Effect of copolymers added (4%) on flexural modulus of the PP/PC (75/25) blends.

increase the strength and the modulus of the materials to a certain extent, more obviously in the PP-enriched blends, but its inclusion did not change the incompatible nature of the system.

The PP/PC (75/25) blend was chosen as a control sample. The effect of different copolymers added in the PP/PC blends in small quantities (4% wt based on the binary blends) on the mechanical properties of materials was observed to compare the

property difference due to the variation of copolymeric species.

The comparison data of the HDT at 1820 kPa are given in Figure 14. The results show that Cop-1 (PP-*g*-St) and Cop-2 (PP-*g*-St/AN) are superior in improving the HDT of the blends to the other copolymers used. Although only 4% was included, the blend with Cop-2 has HDT values 10% higher than those of the control sample. However, the other

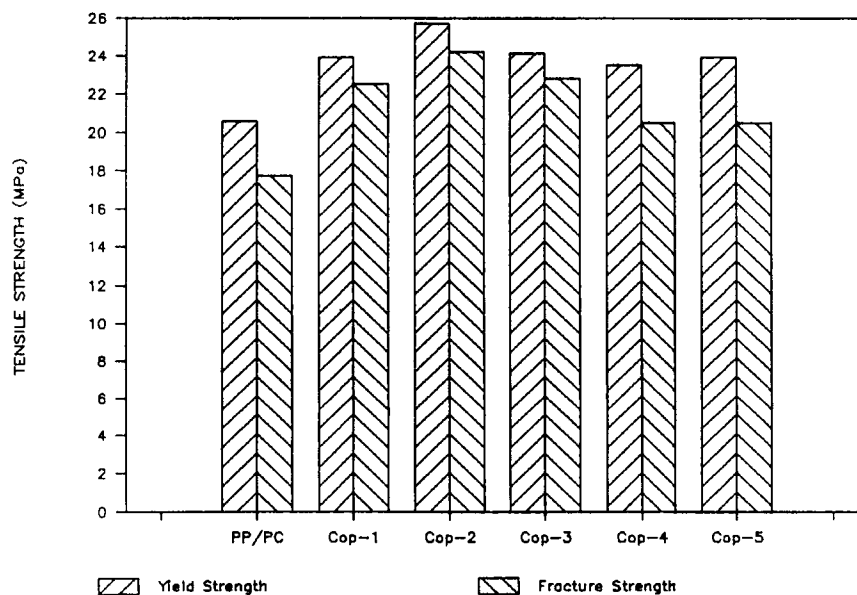


Figure 16 Effect of copolymers added (4%) on the strength of the PP/PC (75/25) blends.

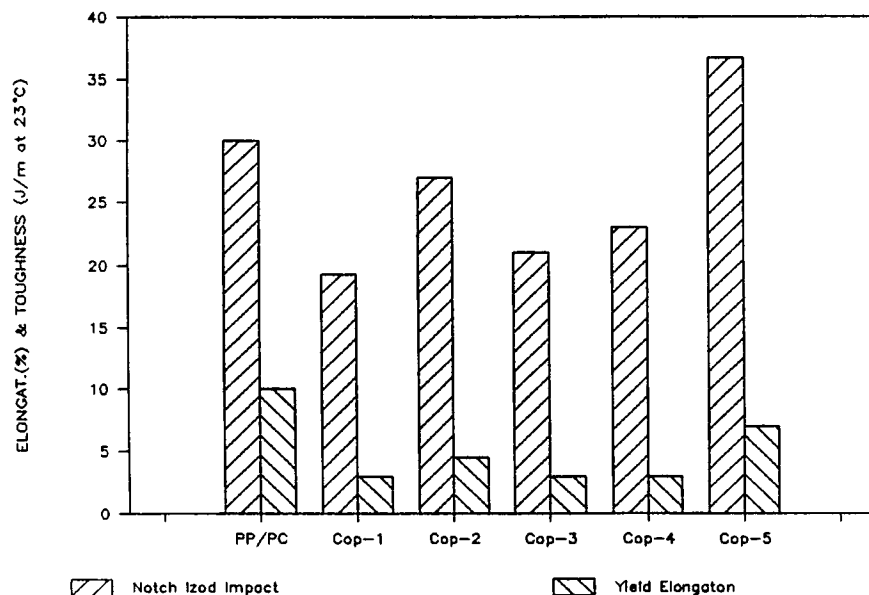


Figure 17 Effect of copolymers added (4%) on the elongation and the toughness of the PP/PC (75/25) blends.

copolymers (Cop-3: PP-*g*-St/BuMA, Cop-4: PP-*b*-BuMA and Cop-5: SEBS) resulted in HDT values lower than those of the control specimen.

The effect of the copolymers added (4% by wt) on the flexural modulus of the PP/PC blends is compared in Figure 15. Although only adding PC could improve the FM of PP material, PP copolymers added to the PP/PC, except SEBS elastomer, increased the FM of the materials to a significantly higher level, particularly using PP-*g*-St/AN as additive.

The effect of copolymers on the tensile property of the PP/PC blends is shown in Figures 16 and 17. The results also suggest that PP-*g*-St/AN (Cop-2), in comparison with other PP copolymers, has a more significant contribution to the improvement of the tensile property of the PP-based blends with PC. Inclusion of Cop-2 (4%) caused the yield strength of the blend to increase by 24.8% and the fracture strength by 36.7%. (see Fig. 16). Although Cop-2 could not bring about an improvement in the ductility properties (such as elongation and impact strength) of the blends, Cop-2 did reduce the ductility of the material to the least extent among the PP copolymers used (see Fig. 17).

SUMMARY

In summary, comparing the above data indicates that Cop-2 (PP-*g*-St/AN) as a minor third com-

ponent provides the greatest contribution to the improvement in mechanical properties for the PP/PC blends. The previous discussions on the other properties in earlier sections support this conclusion. The PP graft copolymer with St/AN chains may not only result in strengthening polar interaction between PP molecular backbone, but, also to some extent, in promoting PP adhesion to the PC phase since the St/AN molecular chains may have good adhesion to PC in some chosen St/AN ratio range²² and the Cop-2's inclusion in the blend could produce the least viscosity reduction and the most significant influence on the degree of undercooling during PP crystallization in the blends. All of those are favorable for such a function of Cop-2 in property improvement, viz. by comparison, the PP/PC/Cop-2 blend having the most favorable balance of properties among the blends studied, such as better endurance of the material at elevated temperature, higher mechanical strength, and modulus and less loss of ductility.

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REFERENCES

1. Z. Dobkowski, Z. Kohman, and B. Krajewski, in *Polymer Blends*, E. Martuscelli, R. Palumbo, and M. Kryszewski, Eds., Plenum Press, New York, 1980.

2. L. Delgiudice et al., U. S. Pat. 4,713,416 (1987).
3. Y. Shinichi et al., Jpn. Pat. 63,105,022, **A2** (1988).
4. T. Megumi et al., Jpn. Pat. 63,215,752, **A2** (1987).
5. M. Takeaki et al., Jpn. Pat. 63,215,714, **A2** (1988).
6. P. Bataille, S. Boisse, and H. P. Schreiber, in *Advances in Polymer Blends and Alloys Technology*, Vol. 1, M. A. Kohudic, Ed., Technomic, Lancaster, PA, 1989, p. 12.
7. A. Rudin and N. E. Brathwaite, *Polym. Eng. Sci.*, **16**, 1312 (1984).
8. B. D. Favis and J. P. Chalifoux, *Polymer*, **29**, 1761 (1988).
9. P. K. C. Pillai, *J. Polym. (Tokyo)*, **16**, 575 (1984).
10. D. W. Bartlett, D. R. Paul, and J. W. Barlow, *Mod. Plast.*, **58**, 60 (1981).
11. V. Dobresu and V. Cobzaru, *J. Polym. Sci. Symp.*, **64**, 27 (1978).
12. D. J. Bye, *J. Eur. Polym.*, **22**, 185 (1986).
13. R. Y. Feng, Y. Y. Lin, W. Luo, and J. Y. Pan, *Zhongshan Daxue Xuebao Ziranhexueban*, **1**, 100 (1983); *Chem. Abstr.*, **99**, 140735n (1983).
14. R. E. Prud'homme and R. S. Stein, *Macromolecules*, **4**, 668 (1987).
15. Overseas Report, *Mod. Plast.*, **April**, 128 (1990).
16. G. Fairley and R. E. Prud'homme, *Polym. Eng. Sci.*, **27**, 20 (1987).
17. B. Z. Jiang, D. R. Uhlmann, and J. B. Vander Sande, *J. Appl. Polym. Sci.*, **29**, 4377 (1984).
18. A. Siegmann, *J. Appl. Polym. Sci.*, **27**, 1053 (1982).
19. A. K. Gupta and S. N. Purwar, *J. Appl. Polym. Sci.*, **29**, 1595 (1984).
20. G. Locati, *Rheol. (Proc. Int. Congr.) 8th*, **3**, 205 (1980).
21. C. E. Locke and D. R. Paul, *J. Appl. Polym. Sci.*, **17**, 2597 (1973).
22. J. W. Barlow and D. R. Paul, *Polym. Eng. Sci.*, **24**, 525 (1984).

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