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Study of the Role of Silane-treated Filler on the Compatibility of Polypropylene/Polystyrene Blends at Different Ratios*

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A systematic study of the effect of the filler on the immiscible blend properties is presented as a function of the blend composition. As a representative of semicrystalline polymer/amorphous polymer blends, a polypropylene/polystyrene system was chosen. The presence of filler in the blends has induced significant changes in the rheological properties. This behavior is a function not only of filler surface treatment but also of blend composition. Solid state dynamic mechanical analysis is performed and the relaxation behavior is correlated with the morphology of the blends. The increase in tensile strength of the blends filled with surface-treated glass beads is related to the enhancement of the adhesion and polymer/polymer interactions.

Keywords: Compatibilization of immiscible blends; polypropylene; polystyrene; silane; coupling agents; filler; dynamic mechanical analysis; polymer melt viscosity

INTRODUCTION

This study is concerned with the compatibilization of immiscible blends by the addition of surface-treated fillers. The primary focus is on the rheological behavior of polystyrene-polypropylene-glass bead

*One of a Collection of papers honoring Yuri S. Lipatov on the occasion of his 70th birthday, 10 July 1997.

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compounds as a function of the filler surface treatment and volume fraction of polymer constituents. A similar study has been reported earlier on a nylon 6-polypropylene system that represents an example of semicrystalline polymer-semicrystalline polymer immiscible blends [1]. An additional system studied in this paper was chosen in order to show more universality of the proposed compatibilization approach by a surface-treated filler. The polypropylene-polystyrene system was chosen as an example of a semicrystalline polymer/amorphous polymer blend.

Polypropylene is one of the most important polymers among the polyolefins due to its intrinsic properties. High melting temperature, high chemical inertness, and low density are some of its most appealing characteristics. The immiscibility of the polypropylene/polystyrene blend has been widely demonstrated, since the blend components have very distinct interfacial tensions in the molten state. Glass beads were chosen as a filler not to optimize properties but to demonstrate the concept of compatibilization by surface-treated fillers by minimizing the rheological complications caused by the aggregate formation of ordinary fillers.

The morphology of immiscible blends depends on the properties of the individual polymer components, their concentration, blending procedure and compatibilization method [2]. Therefore, in order to understand the behavior of a filled immiscible blend, all these factors should be considered. Since the morphology of immiscible blends is a function of composition, the addition of fillers in the blends may change the morphological behaviors in comparison with the non-filled blend. Therefore, the main objective of this study is to understand the relation between rheological and morphological properties with the changes in blend composition and by addition of filler in these blends.

The deviation of the melt viscosity of blends from the rule of linear additivity is related to morphological changes. Cohen *et al.* [3] have discussed the viscosity anomaly of polypropylene/polystyrene blends. They attributed this deviation to interfacial slip. However, for this study, the viscous behavior is more complex since the blend is filled and the filler surface properties are changed by a silane coupling agent. The role of coupling agents is basically to improve filler/matrix interaction [4].

The dynamic viscosity at low frequencies is sensitive to structural changes of the network formed by particles in the polymeric matrix.

All systems are analyzed in terms of complex viscosity. At low shear, the viscosity behavior is governed more by the filler particles than by the suspending medium, whereas the opposite behavior is expected for high-shear viscosity.

In suspensions of glass spheres, the concentration dependence of the viscosity follows the Mooney equation [5]:

$$\ln\left(\frac{\eta}{\eta_0}\right) = 2.5\phi(1 - C\phi) \quad C = 1.351$$

where, η and η_0 are the viscosity of the filled and non-filled systems, respectively, and ϕ is the filler volume fraction.

The addition of an inert solid to a polymer increases the melt viscosity and decreases the melt elasticity. The rheological properties of concentrated suspensions depend on the state of dispersion of the rigid particles. Therefore, the filler treated with a coupling agent can actually lead to a decrease in the filled system's viscosity, because it helps to decrease the interparticle forces, thus preventing the aggregation of particles.

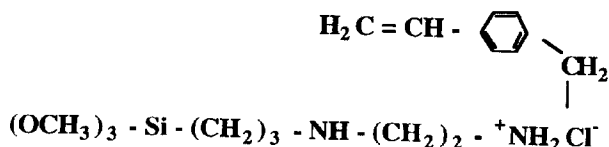
The shear-thinning behavior observed in the filled systems has been attributed to mechanisms in which the shear stress, transmitted through the continuous medium, orients the suspended particles in opposition to the randomizing effects of Brownian motion. Variation of viscosity with shear rate is then a result of the lowered resistance to flow offered by the oriented arrangements. It is seen that, as the loading of filler particles is increased, the melt viscosity increases rapidly at low shear rates. This behavior can be observed better when the viscosity is plotted against shear stress. A proper combination of polymer, coupling agent and filler can lead to drastic changes in the rheological properties of the systems.

The presence of a filler in a polymer matrix usually increases the Young's modulus and decreases tensile strength and elongation, while the presence of rubber decreases the modulus and strength and often increases elongation. As the final properties are of the most interest, the tensile strength of the systems is determined as a function of blend composition and filler surface treatment. The glass transition is also examined, since it is one of the most important variables analyzed in the blend compatibility study.

EXPERIMENTAL

Isotactic polypropylene (PP) was purchased from Aldrich Chemicals Inc. Polystyrene (PS), with a molecular weight of 270,000, was supplied by Dow Chemical. The glass spheres used were supplied by Potters Industries (No. 5000) with a B.E.T. surface area of 0.3 m²/g and average particle size of 10 to 13 μm.

The surface of the glass spheres was chemically modified with a coupling agent, 3-*N*-styryl-methyl-2-aminoethyl aminopropyl trimethoxy-silane, (SAT), purchased from Petrarch Systems Inc. The chemical structure of this coupling agent is shown below.



In order to obtain approximately 10 monolayer equivalence of silane coverage on the filler surface, the amount of coupling agent used was calculated assuming coverage of one molecule of coupling agent to be 0.6 nm². The glass beads were treated with a solution of coupling agent, acidified water which was adjusted to pH 3.5, and ethanol. The water is required for the hydrolysis of the silane. The glass spheres were dried at room temperature overnight with a subsequent heat treatment at 110°C for 10 minutes. The treated and dried glass spheres were sieved to 120 mesh.

The blends were compounded first by dry mixing the polymers with the filler followed by melt blending. The blending was performed in a Banbury mixer equipped with roller-type mixing blades, operated by a Brabender Plasticorder, at 220°C and 80 rpm for 20 min. After mixing, the samples were compression molded at 220°C under 10 MPa of pressure for 15 min, obtaining sheets of 3 mm thickness. The blend compositions are described in Table I where the amount of filler was maintained at 50% by weight.

The morphology of the samples was observed in a scanning electron microscope, Joel JSM-35CF, at an applied voltage of 20 kV. The compression-molded samples were fractured under liquid nitrogen and coated with 9 nm of gold prior to introduction into the microscope.

TABLE I Blend compositions

Blend ratio PP:PS	PP (%weight)	PS (%weight)	Glass beads (%weight)
0:100	0	50	50
20:80	10.2	39.8	50
40:60	20.4	30.6	50
60:40	30.6	20.4	50
80:20	39.8	10.2	50
100:0	50	0	50

The dynamic mechanical measurements were performed in a dynamic mechanical spectrometer, Rheometrics RMS-800, which is equipped with a 2000 g-cm force rebalance transducer. The solid state analysis was performed at a temperature range from -30°C to 150°C , at 0.1 rad/s of frequency, and constant strain 0.1%. The dynamic melt state properties of all compounds was measured using a 25 mm diameter parallel-plate configuration at 200°C . A strain of 10% was used, which permitted the experiments to be performed in the linear viscoelastic region.

Tensile strength was measured by cutting standard dog-bones from the compression molded sheets and testing in an Instron universal testing machine. The cross head speed was set to 5 mm/min at room temperature. The sample dimensions and experimental conditions followed the standard test method described in ASTM D 638M-87b.

RESULTS AND DISCUSSION

Polypropylene

The rheological behavior of the individual polymer blend components is first analyzed to give all the information necessary for the understanding of the rheological behavior of the blend.

Figures 1, 2 and 3 show, respectively, the storage moduli, loss moduli, and $\tan \delta$ of the pure polypropylene and polypropylene filled with silane-treated and non-treated glass beads. Polypropylene is known to exhibit three distinct relaxations [6] in the temperature range of -100 to 130°C . The first relaxation occurs around -60°C

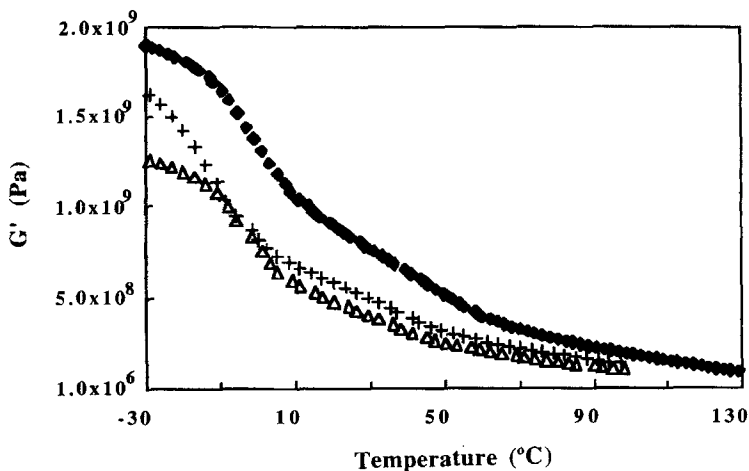


FIGURE 1 Storage moduli of PP (Δ) filled with non-treated (+) and treated (\blacklozenge) glass beads.

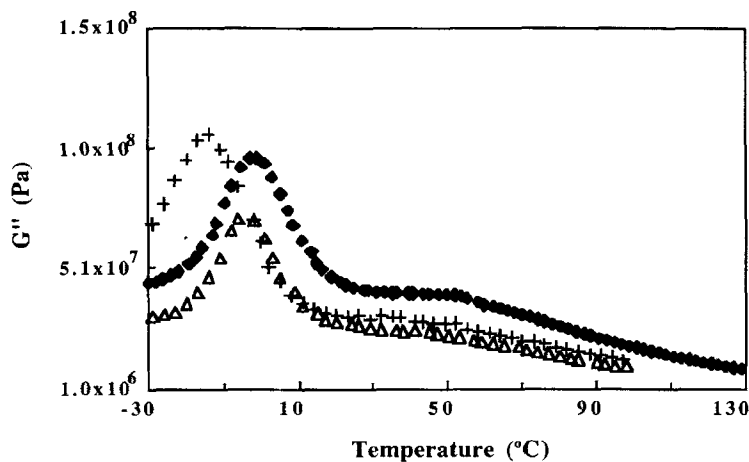


FIGURE 2 Loss moduli of PP (Δ) filled with non-treated (+) and treated (\blacklozenge) glass beads at 50% by weight.

and is usually associated with the γ -transition, which is attributed to a relaxing unit consisting of a few chain segments. The most important transition for the current study is the glass transition, and is designated as β -transition, which appears around 0°C and can be observed in the G'' spectra for the isotactic polypropylene used in this work.

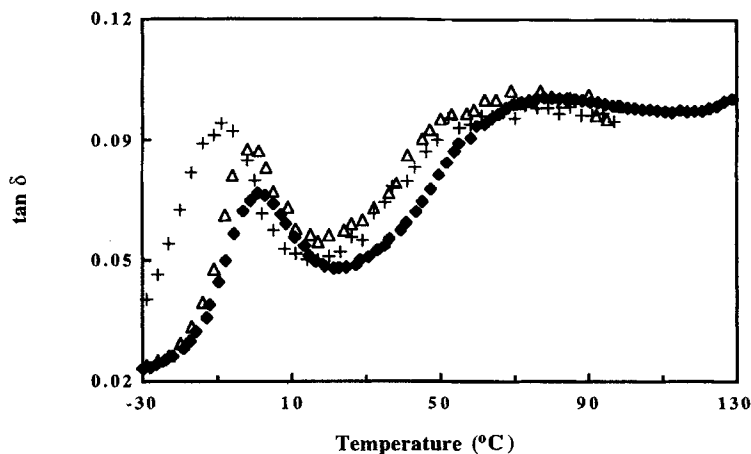


FIGURE 3 Tan δ behavior of PP (Δ) filled with non-treated (+) and treated (\blacklozenge) glass beads at 50% by weight.

The shoulder that appears at 80°C is the α -transition and is associated with the crystalline region. Sumita [7] reported that this transition is present only in highly crystalline polypropylene, *i.e.*, above 20% of crystallinity, and is due to a disorder transition of crystals. Wyckoff [8] found that the smectic crystals transform to the normal monoclinic crystals at 70°C. Therefore the α transition is related to the crystalline region. These transitions are well documented [9] and the obtained results are very similar to those reported in the literature.

The glass-bead-filled PP has distinguishable features in the relaxation behavior [10]. Upon introduction of non-treated filler, the glass transition region in the G'' spectrum becomes broader and more intense. The G'' maximum shifts to lower temperatures in comparison with the pure PP. Such behavior is attributed to the incompatibility of the non-polar PP with the polar surface of the glass beads giving rise to a repulsion effect between both components. Nielsen [11] has attributed the broadening and increase of the transition region in the presence of rigid fillers to the introduction of new damping mechanisms which are not present in the pure polymer. These mechanisms include: particle-particle friction where particles touch each other, particle-polymer friction when there is no adhesion at the interface; and excess damping in the polymer near the interface because of induced

thermal stresses, changes in polymer conformation or morphology. In this specific case, the $\tan \delta$ broadening is mostly attributed to the particle-polymer friction due to the lack of interfacial adhesion as observed in the micrograph of the fractured surface.

On the other hand, an opposite behavior occurs, with lower intensity and shift of the maximum of the glass transition to a higher temperature, upon addition of silane-treated glass beads. The surface treatment enhances the filler/PP interaction, thereby restricting the molecular mobility of PP at the interphase. Changes in the packing density of the polymer chains and modification of the conformation and orientation of chain segments in the interphase are also expected. The α -transition is not as much affected by the presence of filler. However, it is observed that the relaxation starts at a higher temperature when the filler is treated, which is again an indication of the restriction of the molecular mobility.

Polystyrene

The solid state dynamic mechanical spectra of pure and filled polystyrene are shown in Figures 4, 5 and 6. Polystyrene possesses three main relaxations although only the glass transition region is shown here. The δ -relaxation occurs around -240°C and is believed to be

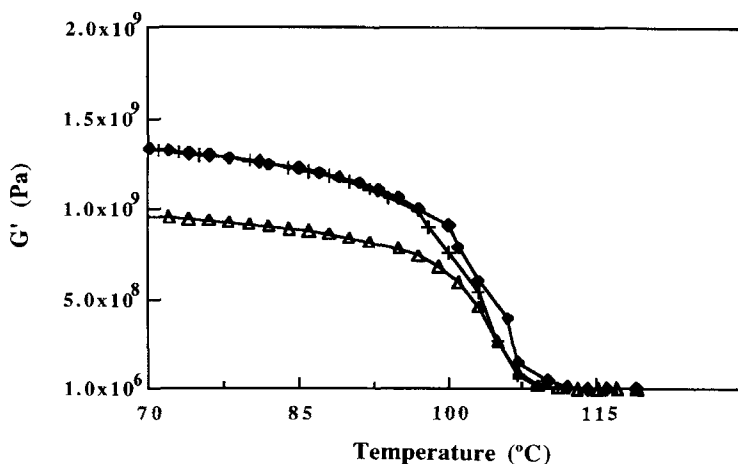


FIGURE 4 Storage moduli of PS (Δ) filled with non-treated (+) and treated (\blacklozenge) glass beads at 50% by weight.

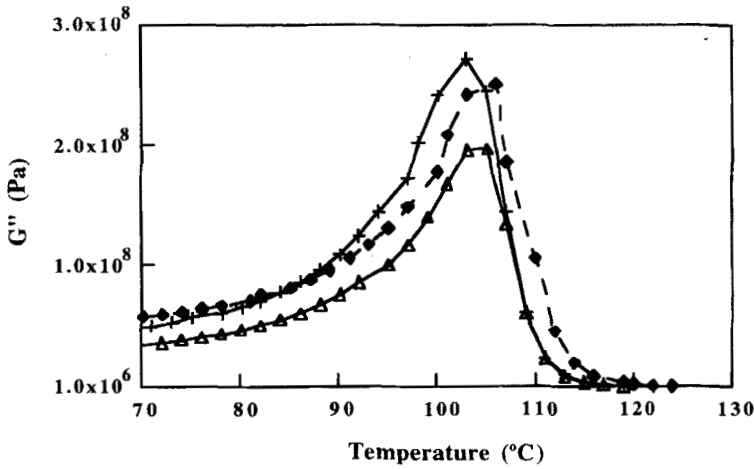


FIGURE 5 Loss moduli of PS (Δ) filled with non-treated (+) and treated (\blacklozenge) glass beads at 50% by weight.

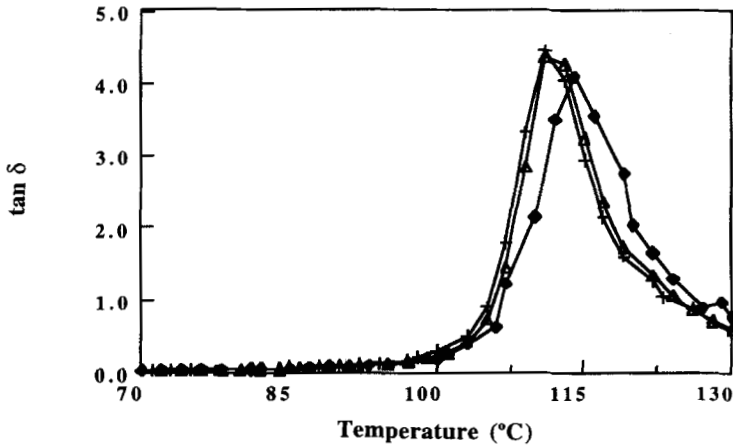


FIGURE 6 $\tan \delta$ behavior of PS (Δ) filled with non-treated (+) and treated (\blacklozenge) glass beads at 50% by weight.

due to the torsional oscillation of the pendent phenyl groups coupled with a wagging motion [12]. Simple rotation of the phenyl groups about their bonds to the backbone is not supported by dielectric or NMR results. The β -relaxation appears as a broad maximum in the $\tan \delta$ spectra at around 35°C, and is due to the phenyl ring reorientation.

The intensity of the $\tan \delta$ peak is sensitive to conformational changes in the polymer backbone. The glass transition occurs around 105°C as shown in Figure 5. After the glass transition, the G'' reflects the molecular weight distribution of the PS. The storage modulus of a high molecular weight material goes through a plateau due to the entanglements before sharply dropping. This drop corresponds to the terminal zone of the viscoelasticity and is attributed to rapid diffusion of polymer chains.

The $\tan \delta$ maximum shifts to higher temperatures after addition of silane-treated filler due to similar reasons as for PP, explained above. However, the simple addition of non-treated filler does not lead to any major changes in the damping behavior. The presence of the filler increased the G' as expected; however, the treatment did not affect the G' value as much.

Polypropylene/polystyrene Blends

The properties of incompatible polymer blends are very complex and have a dependence on the viscoelastic behavior of the individual components, their volume fractions, and the morphology of the blend. The dynamic mechanical results for the non-filled polypropylene/polystyrene blends over the whole range of compositions are shown in Figures 7, 8 and 9, where G' , G'' , and $\tan \delta$ are plotted, respectively, against temperature.

The loss moduli, G'' , are used to determine the glass transition temperatures because they reflect more accurately the relaxation behavior of the polymeric materials. The G'' shown in Figure 8 exhibits two distinctive relaxation peaks corresponding to the individual glass transitions of PP and PS. Due to the complexity and number of spectra to be analyzed, the data are compiled in Figures 10 through 13 to demonstrate major trends of the glass transition and $\tan \delta$ maxima upon compositional changes and filler treatment of the systems.

The behavior of both glass transitions can be better observed in Figures 10 and 11, where the glass transition temperatures of PP and PS are plotted as a function of the blend composition. A noticeable shift of the PS glass transition occurs where the maximum of the G'' changes from 104°C for the pure PS to 108.5°C for the PS in the blends at weight ratios of 6:4 and 4:6 of PP:PS. At a weight ratio of 8:2 of the PP:PS blend, the maximum of the G'' drops to 105°C. This

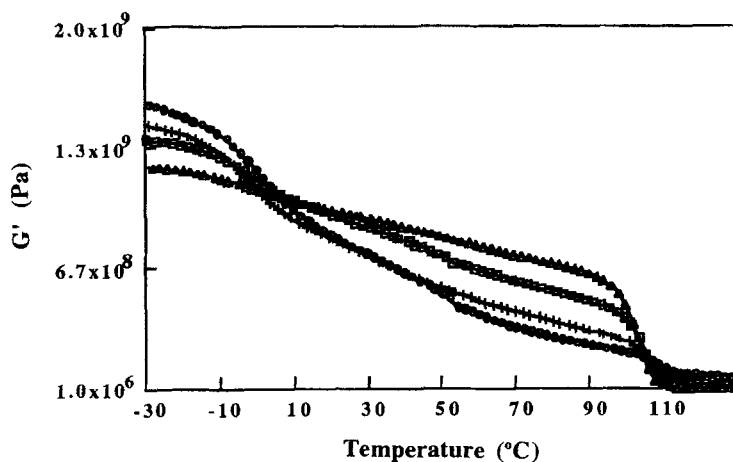


FIGURE 7 Storage moduli of PP:PS at 2:8 (Δ), 4:6 (\circ), 6:4 ($+$) and 8:2 (\square) ratios.

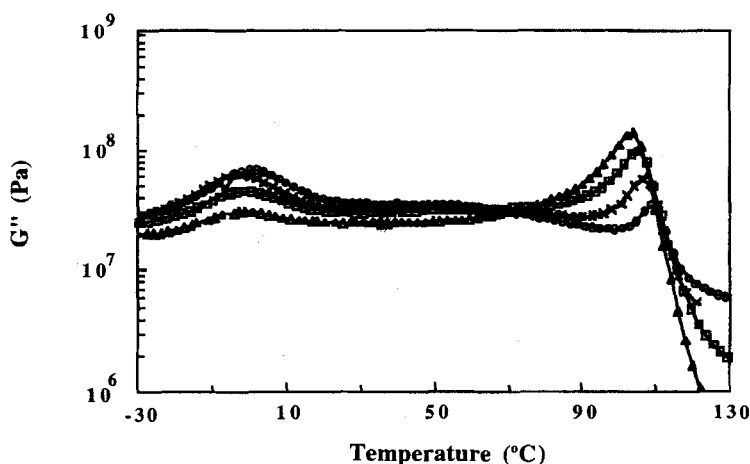


FIGURE 8 Loss moduli of PP:PS blends at 2:8 (Δ) 4:6 (\circ), 6:4 ($+$) and 8:2 (\square) ratios.

is very close to the pure PS transition. The glass transition temperature remains quite constant at around -2°C for PP as shown in Figure 11. Similar arguments as those used previously can be proposed to explain this behavior.

The relative height of the two $\tan \delta$ peaks of the blends is determined by the concentration of the two components and by the morphology of

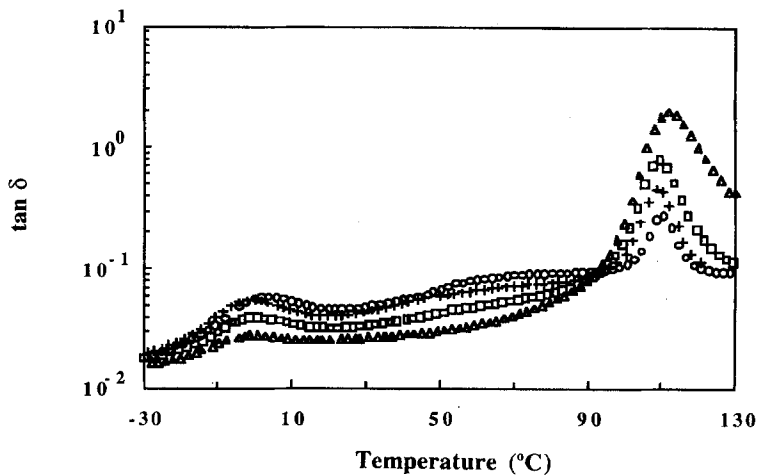


FIGURE 9 Tan δ of PP:PS blends at 2:8 (Δ) 4:6 (\circ), 6:4 (+) and 8:2 (\square) ratios.

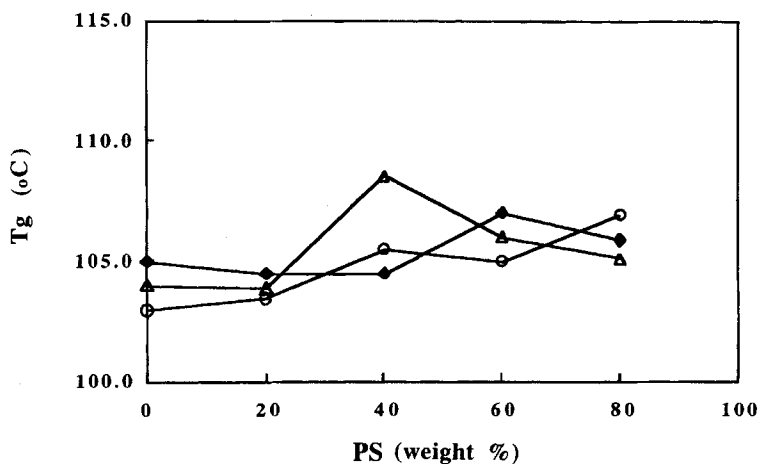


FIGURE 10 Glass transition temperatures of polystyrene (Δ), non-treated (\circ) and treated glass beads (\blacklozenge).

the system. The more continuous the phase is, the higher is its damping peak for the corresponding component. In changing the PP:PS weight ratio of the blends from 2:8 to 8:2 a series of morphological changes are observed in addition to phase inversion. These changes affect the rheological behavior. Figures 12 and 13 present the tan δ

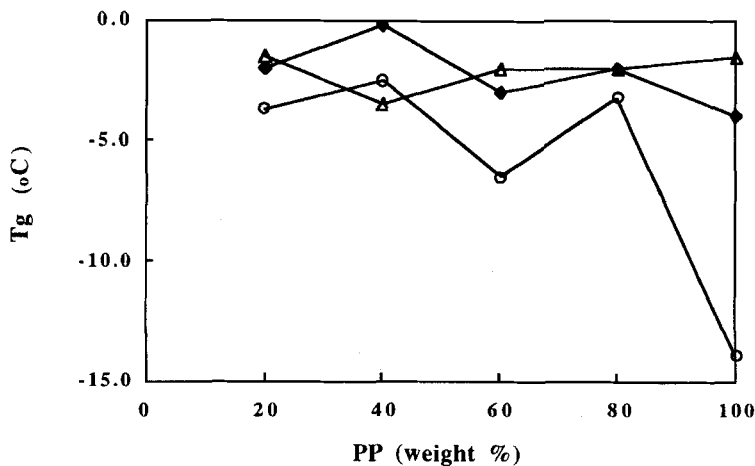


FIGURE 11 Glass transition temperatures of polypropylene (△) filled with non-treated (○) and treated (◆) glass beads.

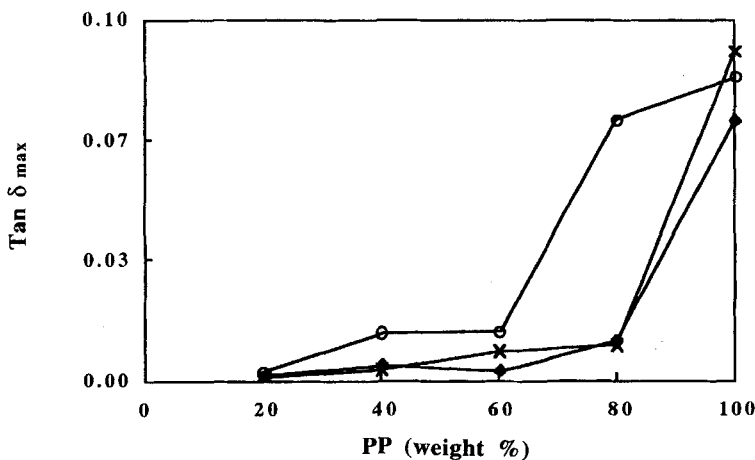


FIGURE 12 Tan δ maximum of PP as a function of PP:PS composition. Non-filled blends (○) and filled with non-treated (+) and treated (◆) glass beads.

maxima as a function of blend composition for PP and PS glass transitions. The tan δ maximum of the PP glass transition increases with PP concentration in the blend as expected, changing from 0.02 for 20% to 0.09 for 100% of PP. However, the PS transition does not follow the same trend, and thus indicates the blends undergo morphological

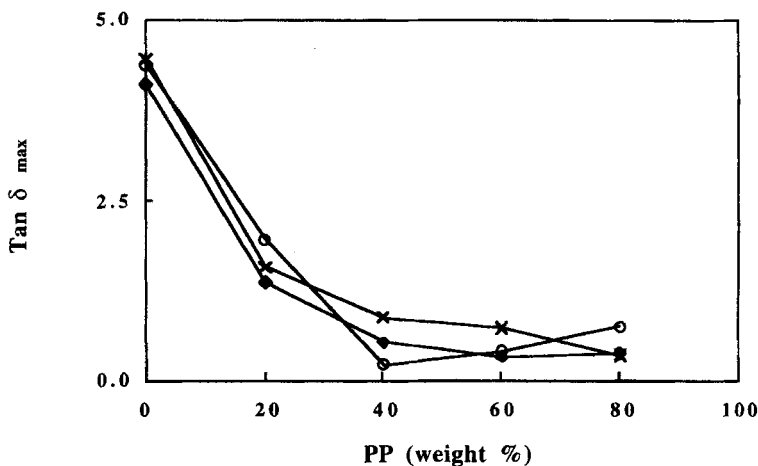


FIGURE 13 Tan δ maximum of PS as a function of PP:PS composition. Non-filled blends (○) and filled with non-treated (+) and treated (◆) glass beads.

changes. The tan δ maximum drops from 4.39 to 0.27 for 100% and 60% of PS, respectively, and increases up to 0.80 for lower PS weight fractions in the blends.

In the region where the viscoelastic functions are slowly varying for both components, one in the glassy region and the other in the rubbery plateau, the additivity of the two phases can be described in terms of a pseudo-equilibrium modulus. Many mixing rules have been described. In general, neither the moduli nor the compliances will be additive [13]. It is clear that the additivity rule is not followed in the blends studied for the two main transitions. According to this rule, the tan δ maximum should be proportional to the volume fraction of the blend components. Analyzing Figure 9, it is clear that the rule is not obeyed. The blends of weight ratio 6:4 and 4:6 of PP:PS show a maximum tan δ intensity for the PP peak, whereas the PS transition of the weight ratio of 4:6 has a less intense peak, followed by the 6:4 weight ratio. This behavior is explained by the fact that around these compositions the phase inversion occurs and the morphology is changed. The phase inversion occurs at the composition where both phases form a continuous phase. The exact composition at which inversion occurs can be changed considerably by the intensity of mixing or by the presence of solvents. Generally, there is a range of

composition where both phases are partially continuous and where the modulus changes very rapidly with the compositions.

The storage moduli behavior of the incompatible blends is illustrated in Figure 7. For compositions 6:4 and especially 4:6 of PP:PS, the moduli shows a deep drop between the glass transitions. Such a change can be due the proximity to the phase inversion. The storage moduli at 60°C are plotted against the whole composition range, as shown in Figure 14, for better visualization. The variation of the moduli of the blends with the composition exhibits a deviation from linearity. The same behavior is observed for temperatures below the PP glass transition.

Polypropylene/polystyrene Blends Filled with Glass Beads

In order to perform a systematic analysis of the role of the filler in PP:PS blends, the blend compositions were varied and silane-treated and non-treated glass beads were added in each blend.

An interesting change in the phase inversion behavior occurs upon addition of non-treated glass beads and further with treated glass beads. Upon addition of filler in the blends the drop of the moduli between the glass transitions occurs at different compositions. The

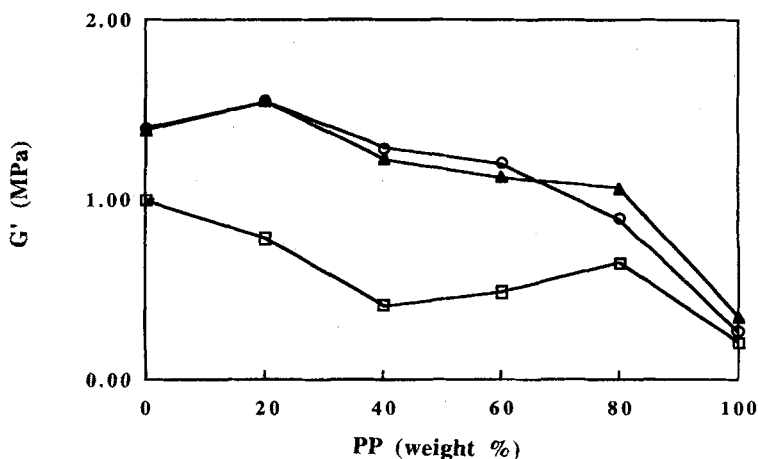


FIGURE 14 Storage moduli of PP:PS blends (□) filled with non-treat (○) and treated (▲) glass beads at 60 °C.

decrease of G' is observed for the 6:4 and 4:6 ratios of PP:PS for the non-filled blends. Upon adding non-treated glass beads, this drop shifts to the weight ratios of 6:4 and 8:2 of PP:PS. Finally, for the addition of silane-treated glass beads, it occurs only for the 8:2 PP:PS weight ratio. This behavior is quite interesting, showing either a change in the phase inversion composition or a change in morphology without phase inversion. As shown in Figures 10 and 11, the PP glass transition increases 1 or 2°C in comparison with the non-treated filler as an overall behavior. This increase is too small to assign any structural changes to this behavior. The glass transitions shift toward each other upon compatibilization for miscible blends; however, the majority of the publications about compatibilization of immiscible blends do not observe such a shift even though an enhancement of the compatibilization is achieved. Utracki [14] presented similar results for polypropylene–Nylon 6 blends where the compatibility had affected the Nylon 6 more than the dispersed phase, PP. In our study, the PP and PS glass transitions increase upon filler surface treatment as expected. The addition of treated filler brings the glass transitions closer to each other in comparison with the non-filled blends and even with the filled blends with non-treated glass beads. Consequently, an increase of the glass transition is observed due to enhancement of the matrix/filler interaction and also a shift toward each other up to 3°C.

The glass transition is characterized not only by the G'' maximum temperature but by its range and intensity. Therefore, the above analysis is not conclusive without further examination of the $\tan \delta$ intensity and shape. The $\tan \delta$ of PP, shown in Figure 12, has gone through a substantial decrease upon addition of non-treated filler, which can be due to the decrease of the PP concentration. However, the filler treatment promoted a further decrease in the $\tan \delta$ maxima which can be seen for both PP and PS. Besides the decrease of the $\tan \delta$ maximum, the transition has broadened, mainly for the composition 6:4 and 4:6 of PP:PS, as shown in Figures 15 and 16. The broadening is attributed to the enhancement of molecular intermixing.

Viscosity

The viscosity results are plotted in Figures 17 through 22, where for each composition three spectra are shown for the non-filled polymeric

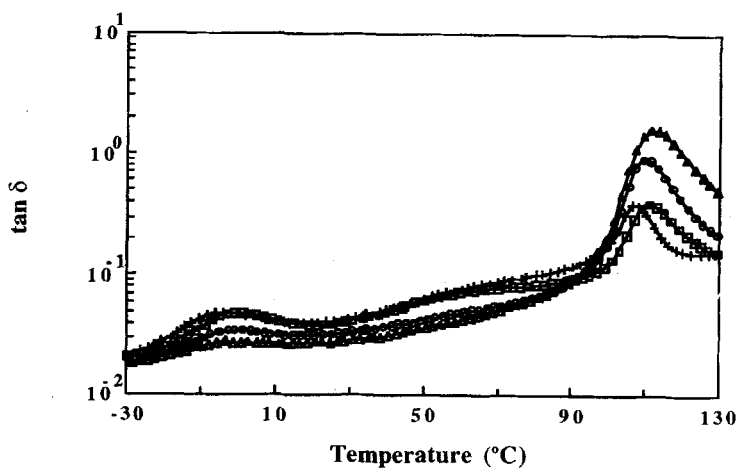


FIGURE 15 $\tan \delta$ of PP:PS blends at 2:8 (Δ), 4:6 (\circ), 6:4 (+) and 8:2 (\square) ratios filled with non-treated glass beads.

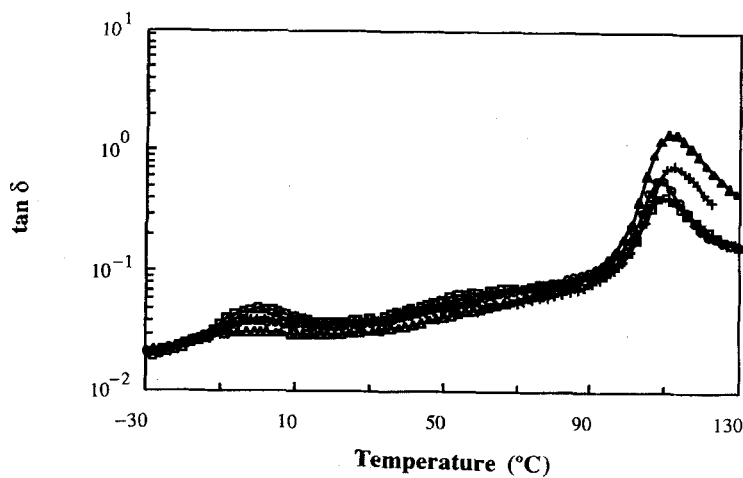


FIGURE 16 $\tan \delta$ of PP:PS blends at 2:8 (Δ), 4:6 (\circ), 6:4 (+) and 8:2 (\square) ratios filled with treated glass beads.

system and blends filled with non-treated and treated glass beads. Figures 21 and 22 reflect a well-known behavior where the viscosities of the pure components PP and PS are the lowest, followed by the pure polymeric components filled with the treated filler. The coupling

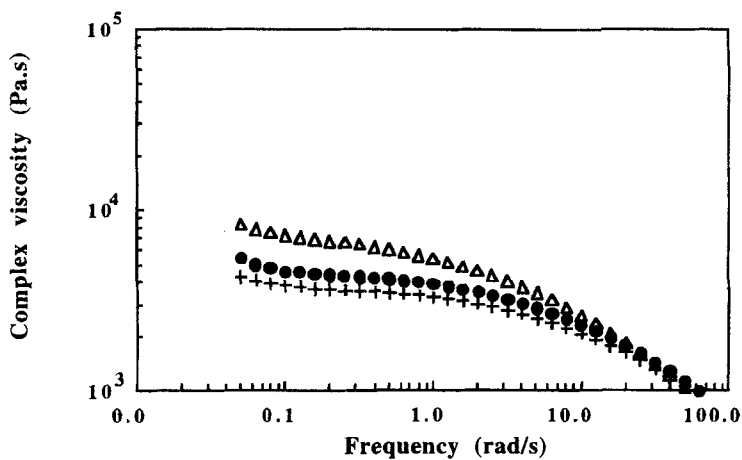


FIGURE 17 Complex viscosity of PS (Δ), PS filled with non-treated (+) and SAT treat (\bullet) glass beads.

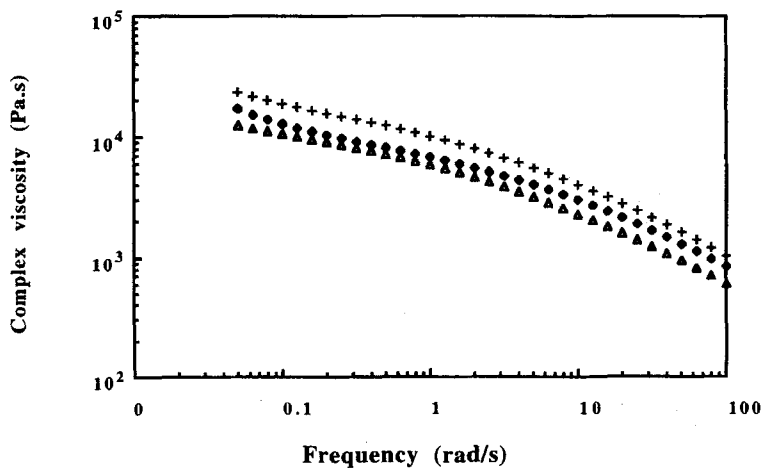


FIGURE 18 Complex viscosity of PP/PS blend (Δ) at 2:8 ratio filled with non-treated glass beads (+) and SAT (\blacklozenge) treated glass beads.

agent enhanced the filler dispersion, decreasing the viscosity of the polymer filled with the non-treated filler.

The melt behavior of filled polymers has been widely studied, as well as the effect of filler surface treatment on these properties. A

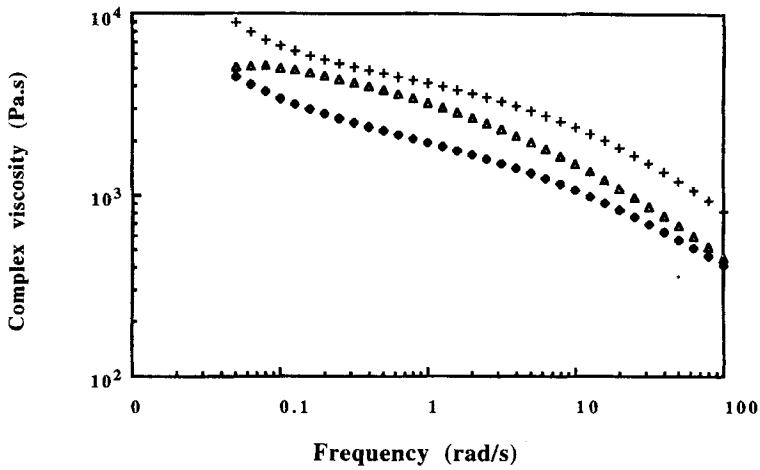


FIGURE 19 Complex viscosity of PP/PS blend (Δ) at 4:6 ratio filled with non-treated glass beads (+) and SAT (\blacklozenge) treated glass beads.

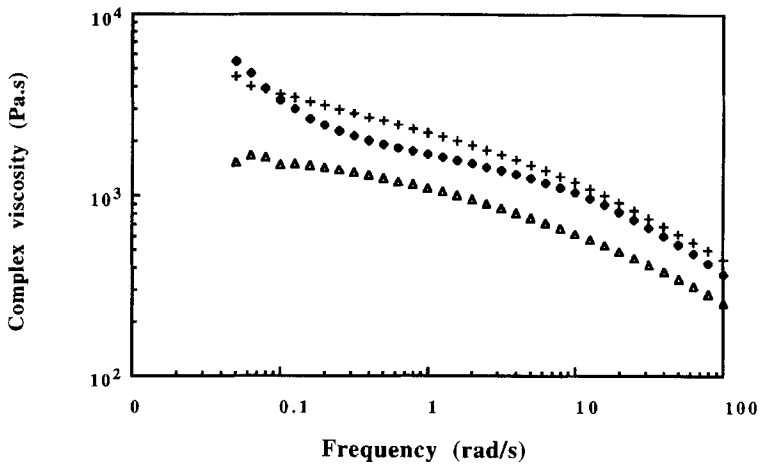


FIGURE 20 Complex viscosity of PP/PS blend (Δ) at 6:4 ratio filled with non-treated glass beads (+) and SAT (\blacklozenge) treated glass beads.

general trend has been observed from these studies, which is the decrease of the viscosity upon filler surface treatment in comparison with the non-treated filler, due to decrease of interparticle forces. Han [15] has been quite reluctant to accept such an explanation because it

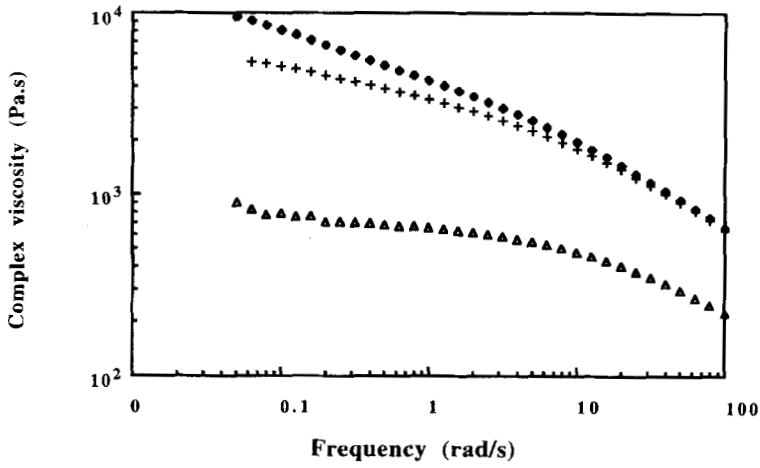


FIGURE 21 Complex viscosity of PP/PS blend (Δ) at 8:2 ratio filled with non-treated glass beads (+) and SAT (\blacklozenge) treated glass beads.

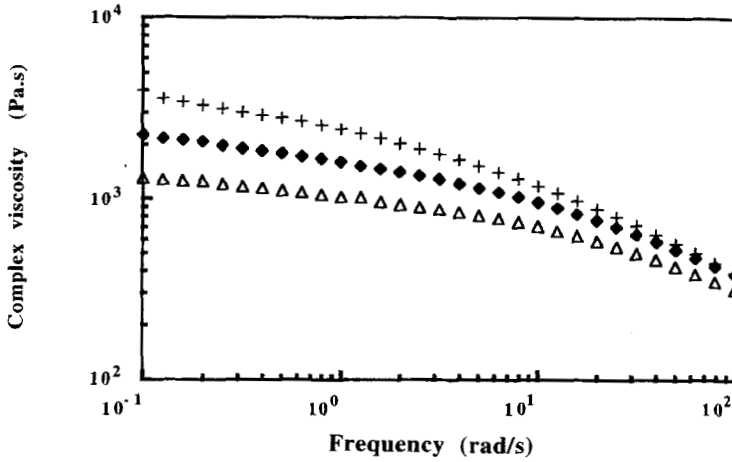


FIGURE 22 Complex viscosity of PP (Δ) filled with non-treated glass beads (+) and SAT (\blacklozenge) treated glass beads.

is difficult to imagine how a reduction of melt viscosity can occur if macromolecules are interconnected to each other through filler particles, when a coupling agent acts as a true coupling agent. The same way of thinking is used in the proposed mechanism, *i.e.*, as the macromolecules

of PP and PS are interconnected at the filler surface due to filler surface treatment an increase of melt viscosity should be expected.

Figure 23 illustrates the relationship between the complex viscosity and the blend compositions, where the viscosity values were obtained at a frequency of 0.1 Hz. The shape of the curves can be an indication of the morphology of the systems and phase interaction. In reality there is a diversity of viscosity-composition curves observed which can be summarized as: viscosity decreases or increases monotonically, the viscosity curve shows an S-shape with blend with blend composition, or viscosity goes through a minimum or maximum. All these curves depend on the mode of dispersion, which depends on the blending ratio and processing conditions. Since the processing conditions and filler content were held constant, the blending ratio and also the filler surface treatment must be responsible for the morphological changes.

The negative deviation from linearity can be attributed to the lack of adhesion between the two phases. A positive deviation indicates that strong interaction between both polymer components is achieved upon compatibilization with graft copolymer. The plot of blend viscosity-composition shows a negative deviation from linearity at a blend composition of 8:2 for the PP:PS system. The S-shape of this curve is

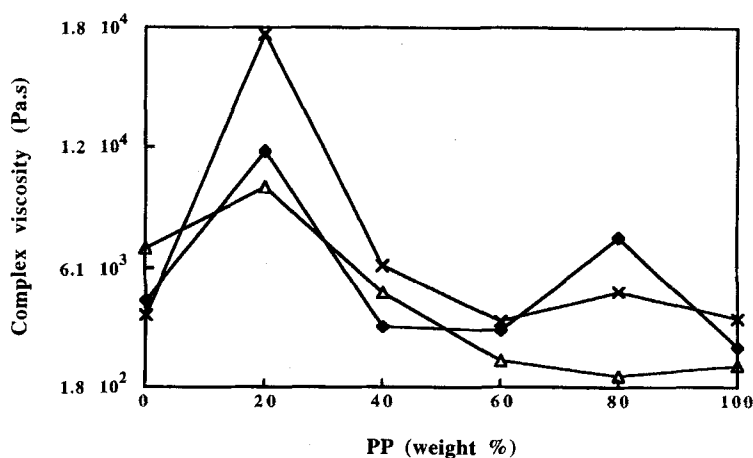


FIGURE 23 Complex viscosity of the non-filled (Δ), and filled blends with non-treated (+) and treated (\blacklozenge) glass beads as a function of PP weight %.

attributed to the poor interaction among the system components [16]. The positive deviation from linearity at all compositions for the blends filled with treated glass beads is attributed to the interactions among the phases, as observed in Figures 24 and 27. At the 6:4 composition of PP:PS, a more interlocking morphology is present, while at a 2:8 composition a droplet-like morphology is seen. The viscosity behavior can be attributed to the morphological changes. The maximum of the curve for the blends with the non-treated filler at 2:8 PP:PS is due to the elongated droplet-like morphology in comparison with the non-filled blend as shown in Figures 25 and 24, respectively.

The filler surface treatment enhances the matrix/filler interaction, decreasing the phase separation size and shape, as seen in Figure 26. The change in morphology reduced the viscosity; however, this composition shows a maximum in the curve of the blends filled with treated filler. Therefore, the lower viscosity does not mean a weaker interaction between the components but indicates a different morphology.

The 4:6 composition shows a behavior similar to that for the 2:8 PP:PS weight ratio. The viscosity of the systems filled with treated filler starts to increase at a weight ratio 6:4, where a more interlocking

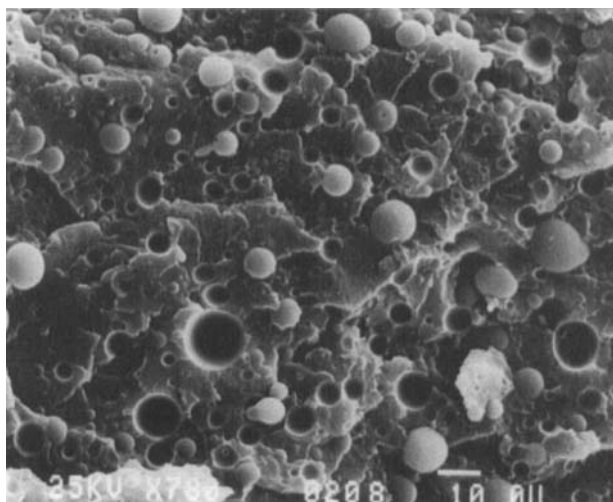


FIGURE 24 Micrograph of the polypropylene-polystyrene blend at ratio 2:8.

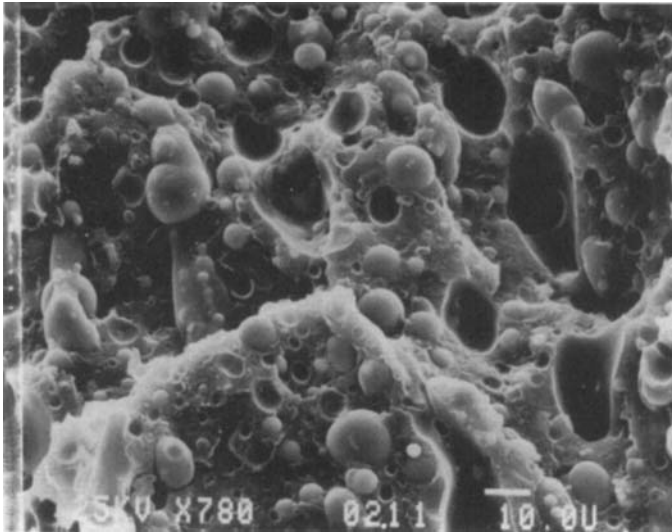


FIGURE 25 Micrograph of the polypropylene-polystyrene blend at ratio 2:8 filled with glass beads.

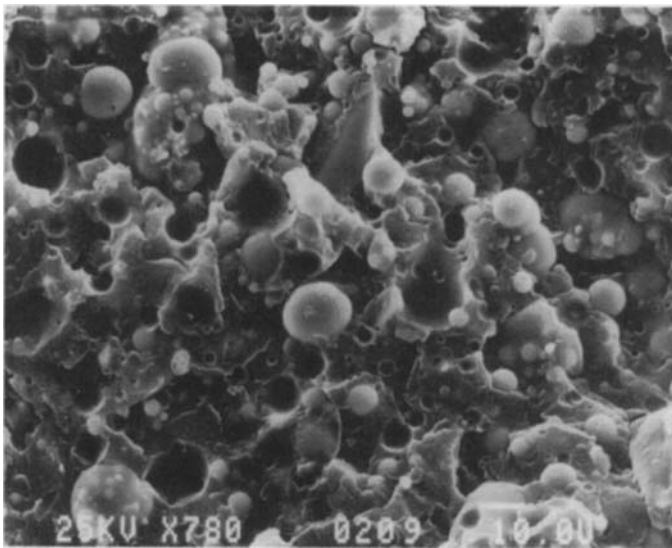


FIGURE 26 Micrograph of the polypropylene-polystyrene blend at ratio 2:8 filled with surface treated glass beads.

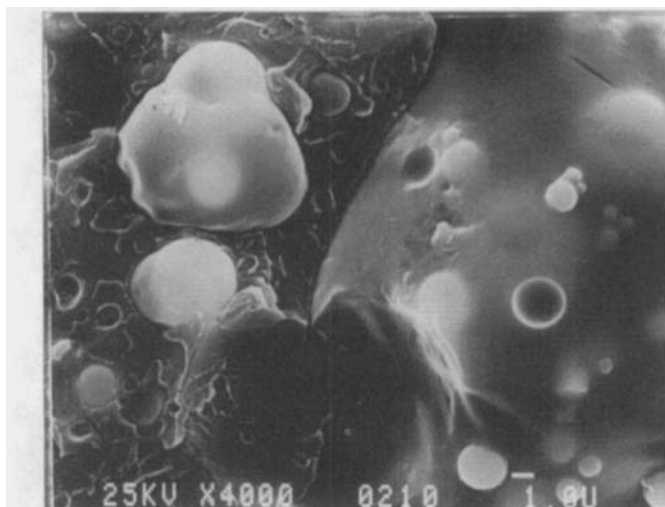


FIGURE 27 Micrograph of the polypropylene-polystyrene blend at ratio 2:8 filled with non-treated glass beads.

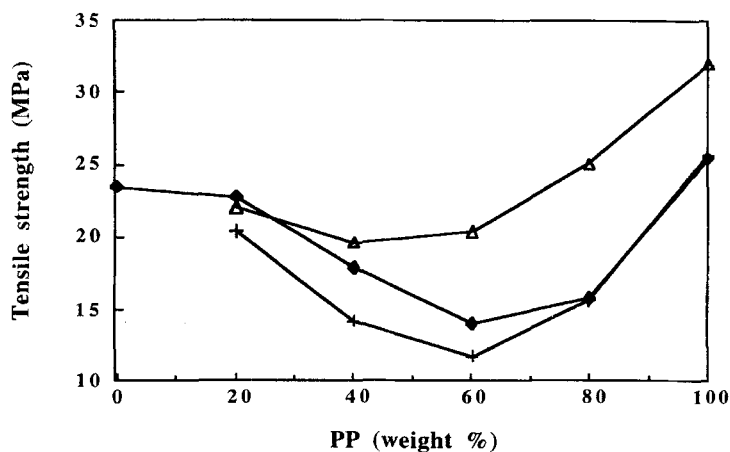


FIGURE 28 Tensile strength of PP:PS blends as a function of composition and filler surface treatment. Blend (△), filled with non-treated (+) and treated glass beads (◆).

morphology is observed. Actually, at very small shear rates the viscosity of the 6:4 PP:PS weight ratio blend filled with treated filler is higher than that of the blend filled with non-treated filler.

It is interesting to mention the morphology shown in Figure 27, where a detachment of a glass bead from a PP:PS interface is observed. If the adhesion between PP/glass bead and PS/glass bead was adequate in that area, an efficient stress transfer among the three phases would have occurred, as proposed by the interphase compatibilization mechanism.

Tensile Strength

The influence of the filler and coupling agent on the mechanical properties of filled systems was examined. It was determined that the modulus of elasticity of an incompatible thermoplastic polymer blend follows an *S*-shape curve if plotted against composition [17]. The fracture path of polystyrene/polypropylene blend is expected to follow the weak interface between the polymer phases or that fracture is initiated at the interface. However, the tensile strength does not follow such a curve but shows a minimum around 5:5 compositions, as shown in Figure 28. The decrease in tensile strength of PS and PP upon blending can be attributed to several causes. A poor interaction exists between both polymers and stress concentration is created around the inclusions. The tensile strength response for filled systems has shown various behaviors, increasing or decreasing as a function of the interfacial properties. For example, the coupling-agent-treated glass beads have shown a better enhancement of the PS tensile strength than that of the PP. The tensile strength of the filled blends increases upon filler treatment in comparison with the non-treated filler. Such behavior can be understood if the coupling agent is improving the adhesion among the three phases, as shown in the micrographs. Upon treating the filler surface, dispersion and surface wetting are improved, therefore the state of dispersion and the interface characteristics affect the tensile strength of the systems. Two exceptions are observed. Filled PP and filled 8:2 PP:PS compositions do not present major changes.

CONCLUSIONS

The multicomponent polypropylene/polystyrene/glass bead system showed a strong influence of the morphology on the rheological and

mechanical properties as a function of blend composition. The filler surface treatment alters the polymer/filler and polymer/polymer interface at the filler surface. This leads to a completely different morphology as compared with the non-filled blends. The melt state viscosity is not only a function of the blend composition, but also a function of the morphological change induced by the filler and its surface treatment. The tensile strength of the system is enhanced over the entire range of compositions upon addition of surface-treated filler, which is explained by the enhancements of the interfacial interaction among the three phases.

Therefore, an adequate surface treatment can bring an enhancement of the compatibility of immiscible blends. The proposed compatibilization mechanism is effective for a semicrystalline/amorphous blend.

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