Blends of Polypropylene with Solid Silicone Additive

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ABSTRACT: Blends of polypropylene (PP) and silicone masterbatch (SMB_{PP})-a commercial formulation consisting of an ultrahigh molecular weigh polydimethylsiloxane (PDMS) dispersed in PP-were prepared by melt mixing in an internal mixer. Four binary blends with different SMB_{PP} content and two ternary blends containing silanegrafted polypropylenes (PP-VTES) of different VTES content as compatibilizer were produced. The blends were analyzed by melting flow rate, rotational rheometry, scanning electron microscopy, and differential scanning calorimetry (DSC). In all blends, the SMB_{PP} remained as dispersed phase in the continuous PP matrix. The addition of PP-VTES reduced significantly the size of the SMB_{PP} domains. Rheological and morphological data strongly indicate that the PP-VTES acts only by lowering the interfacial tension of the system without generating strong

INTRODUCTION

Polyolefins are the largest class of commercial polymers due to their low price and good mechanical properties, and are widely used as blends with one another and other classes of polymers. Polysiloxanes are among the materials having the potential to be used in blends with polyolefins. Polydimethylsiloxane (PDMS) is the most widely used polysiloxane for this type of application because of its well-known surface-modifying properties.¹ Its structure is composed of highly flexible Si-O bonds in the main chain, with methyl groups attached to the silicon atoms¹; so its physical and chemical properties combine both inorganic and organic characteristics. The surface properties of PDMS are very attractive for modification of other surfaces and interfaces. However, its low solubility parameter makes it highly immiscible with a variety of organic polymers. This

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interaction between SMB_{PP} domains and the PP matrix. The dimensions of the SMB_{PP} domains and the interfacial energy were observed to determine the characteristics of the pseudoplastic behavior of the blends in the melt state. For both binary and ternary blends, the SMB_{PP} domains showed nucleating effect leading to an increase of the degree of crystallinity. However, the decrease in the interfacial energy and viscosity promoted by the addition of PP-VTES to the system led to a more intense nucleating effect and to an increase of crystallization, melting temperatures, and melting enthalpy. © 2007 Wiley Periodicals, Inc. J Appl Polym Sci 104: 226–233, 2007

Key words: polypropylene; blends; polydimethylsiloxane; silane-grafted polypropylenes; thermal and rheological properties; blend compatibilization

immiscibility leads to PDMS rejection from the matrix when used in blends with other polymers.

Due to the fact that the addition of PDMS to polyolefins can lead to advantages in processing and improvements in the surface properties with low adverse effect on mechanical properties, polyolefin/ PDMS blends have been the subject of many studies in the last years.²⁻¹² Falender et al.³ studied blends of PDMS with ethylene-vinyl acetate (EVA) copolymers under shearing and high temperature conditions. When compared to pure polyethylene, these blends showed lower modulus over the temperature range of -150 to $+ 65^{\circ}$ C and lower mixing energies. The electrical properties showed superior or intermediate values in comparison with values observed in the pure components. Santra et al.⁴ studied the effect of the addition of ethylene-methacrylate (EMA) copolymer to 50/50 blends of low-density polyethylene (LDPE) and PDMS rubber, showing that EMA acted as a very good compatibilizer in these blends. The optimum level of EMA compatibilizer for the blend was reported to be 6 wt %. Bhattacharya et al.⁵ have reported preliminary studies on the rheological behavior of EMA/PDMS blends at various temperatures and different shear rates. They found that the blends presented positive deviation with relation to the additivity rule, indicating the presence of syner-

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gism. Jana et al.^{6,7} achieved the compatibilization of LDPE/PDMS blends through reactive processing in the presence of EMA. Jana and Nando⁸ also showed that the compatibilized LDPE/PDMS blends could be used as effective cable insulants. Yilgor et al.⁹ studied the effect of small amounts of silicone-containing block copolymers on polypropylene (PP) and high-density polyethylene (HDPE) through blending. The extrusion rates of these blends were improved and surface properties were modified. Bulk properties of these modified systems, such as crystallization, melting behavior, and tensile properties, were not affected. Jana and Nando¹⁰ studied the effect of different ethylene copolymers as compatibilizers on 75/25 LDPE/PDMS blends, having found that EMA, EVA, ethylene acrylic acid (EAA) and an ionomer (Lotek-4200) were effective as compatibilizers. In addition, the authors reported that the compatibilizers improved physical properties and thermal stability of the blends, decreased the size of the phase domains and influenced dynamic properties of the blends. Recently, Jana and Nando¹¹ studied the rheological behavior of LDPE/PDMS rubber blend at different temperatures and shear rates with varying proportions of EMA copolymer and silica filler loading. They found that all blends were pseudoplastic in nature and that both PDMS and EMA compatibilizer contents presented effect on the rheological behavior of the blends, affecting their flow behavior index and activation energy. Muñoz et al.¹² studied blends of HDPE with silicone masterbatch (SMB), which are pelletized micro-dispersions of ultra-high molecular weight PDMS in polyolefin carrier resins at loadings of up to 50%,13 and silane-grafted polyethylene (HDPE-VTES). These authors reported that the addition of the SMB improved the HDPE processability with only a slight lowering of mechanical properties. Besides that, the HDPE-VTES used as compatibilizer showed noticeable effect on the size of the SMB domains, which was found to be a determinant factor of the final degree of crystallinity of those blends.

Organosilanes are a class of reactive compounds that find many applications as crosslinking agents for polyolefins,^{14,15} as coupling agents for glass fiber or wood derivatives polymer composites,^{15,16} and as compatibilizer for polymer blends.^{12,17} Vinylalcoxysilanes can be grafted onto polyolefin backbones by the use of small amount of peroxide.^{15,18} Subsequent reactions are generally described as proceeding through hydrolysis of the alcoxyl groups to hydroxyl groups.

In this study, blends of PP and silicone masterbatch MB50-001 (a silicone masterbatch containing 50% of ultra high molecular weight PDMS dispersed in PP¹³) were prepared by melt mixing to analyze the influence of the SMB on the processing, thermal, and rheological properties of PP. The effect of addition of PP-VTES with different VTES levels to the PP/silicone blends was also analyzed.

EXPERIMENTAL

Materials

Additived PP, density 0.9 g/cm^3 and melt flow rate 22 g/10 min, was supplied by Ipiranga Petroquímica S.A. (Triunfo, Brazil). Silicone masterbatch (MB50-001, containing 50% of ultra high molecular weight PDMS dispersed in poplypropylene, referred to in this study as SMB_{PP}) was obtained from Dow Corning Co. (Midland, MI). The PP-VTES compatibilizers were synthesized in our laboratories.¹⁸

Preparation of the blends

PP, PP-VTES, and SMB_{PP} (all predried overnight) were melt mixed at 170°C in a Haake Rheomix 600 internal mixer with rotor speed of 60 rpm for 10 min. After being removed from mixer chamber, the blends were immediately pressed at 170°C for 20 s to obtain thin sheets and, finally, cut into small pieces for tests.

Analyses

The melting flow rates (MFR) of PP and PP/SMB_{PP}, PP/PP-VTES/SMB_{PP} blends were measured using a plastometer, under a load of 2.16 kg at 230° C (ASTM D1238).

Differential scanning calorimetry (DSC) was used to analyze the thermal behavior of the polymers. TA Instruments DSC 2920 and normal calibration procedures involving standard material were used. Fiveto-ten milligram samples were encapsulated in aluminum pans and heated or cooled at 10°C/min under nitrogen. Before any measurement, each sample was heated up to 200°C and maintained at this temperature for 5 min to erase their thermal history. The degree of crystallinity of blends was evaluated by the ratio between the melting enthalpy of the blend and the melting enthalpy of the perfectly crystalline PP ($\Delta H_m^\circ = 50 \text{ cal/g}$).¹⁹

The morphology of pure PP, PP/SMB_{PP}, and PP/PP-VTES/SMB_{PP} blends was studied using a scanning electron microscope (JEOL JSM 5800). Samples were fractured under liquid nitrogen, and the fractured surfaces were coated with gold.

The melt rheology of pure PP, PP/SMB_{PP}, and PP/PP-VTES/SMB_{PP} blends was analyzed in a controlled strain rheometer ARES, from Rheometrics Scientific. Frequency sweeps from 0.01 to 500 rad/s were performed using 25 mm diameter parallel plates at 170° C.

TABLE I Total Results of PP, SMB_{PP}, and Blends

<u>c</u> 1	PP-VTFS	() (D						
code	(wt %)	SMB _{PP} (wt %)	Final torque (Nm)	MFR (g/10 min)	Melting temperature (°C)	ΔH_m (cal/g)	Crystallization temperature (°C)	Crystallinity (%)
PP	_	_	3.0	22	161	21.5	113	43
SMB_{PP}	_	100	-	_	162	11.5	115	23
B _{0/2}	_	2	2.8	24	161	21.2	112	43
B _{0/5}	_	5	2.9	21	161	21.0	113	44
B _{0/10}	_	10	2.9	20	161	20.3	113	45
B _{0/20}	_	20	3.0	21	160	19.3	113	48
$B_{5(2.81)/20}$	5	20	2.5	26	164	19.9	119	50
B _{5(6.07)/20}	5	20	2.7	38	164	20.0	121	50

 ΔH_m = melting enthalpy.

RESULTS AND DISCUSSION

The blends were designated as $B_{x(w)/y}$, where *x* and *y* are numbers indicating the respective contents of PP-VTES and SMB_{PP} in the blends. The subindex (*w*), used only for blends containing PP-VTES, indicates the VTES content in the PP-VTES expressed in weight percent. Then, for example, $B_{0/5}$ refers to a binary PP/SMB_{PP} blend with 5% of SMB_{PP}, and $B_{5(6.07)/20}$ represents a PP/PP-VTES_(6.07 wt % of VTES)/SMB_{PP} blend with a weight composition of 75/ $5_{(6.07)}/20$. The total results of pure components and blends are shown in Table I.

Figure 1 shows the torque–time curves of pure PP and its blends. For all samples, torque values became constant after about 3 min, indicating absence of degradation processes in the system. The torque–time curves were also used to determine the final torque values reported in Table I, which were obtained as the mean of the torque values at the last 3 min of measurement for each sample. For the binary blends, the final torque values were very similar to that of pure PP (2.8–3.0 Nm), regardless of the SMB_{PP} content of the blend. The blends containing PP-VTES



Figure 1 Torque–time curves for pure PP (—) and PP/PP–VTES/SMB_{PP} blends: 98/0/2 (—), 95/0/5 (—), 90/0/10 (—), 80/0/20 (—), 75/5(2.81)/20 (—+), 75/5(6.07)/20 (—×—).

showed somewhat lower final torques (2.5–2.7 Nm). This result is in agreement with the dynamic viscosity curves (Fig. 2) and the MFR values (Table I), which show that the ternary blends presented the lowest viscosity values. This decrease of viscosity in the ternary blends can be attributed to the presence of the PP-VTES, given that the PP grafting process leads to a significant decrease in the molecular weight.¹⁸

Considering the correspondence between steady shear viscosity and dynamic viscosity given by the Cox–Merz rule²⁰ ($\eta^*(\omega)|_{\omega\to 0} = \eta(\dot{\gamma})|_{\dot{\gamma}\to 0}$), the Carreau–Yasuda viscosity model²¹ was used to fit the experimental data from Figure 2, which presented standard deviations of the order of 5%. The general equation for this model is the following:

$$\frac{\eta - \eta_{\infty}}{\eta_0 - \eta_{\infty}} = \left[1 + (\lambda \dot{\gamma})^a\right]^{(n-1)/a} \tag{1}$$

where η , η_0 , and η_∞ are the predicted, the zeroshear-rate, and infinite-shear-rate viscosities, respectively, while the parameter *a* describes the sharpness of the transition from the Newtonian to shear thin-



Figure 2 Dynamic viscosity curves for pure PP (—) and PP/PP–VTES/SMB_{PP} blends at 170°C: 98/0/2 (———), 95/0/5 (——), 90/0/10 (——), 80/0/20 (——), 75/5(2.81)/20 (—+—), 75/5(6.07)/20 (—×—).

п
$.378 \pm 0.020$
$.108 \pm 0.075$
$.219 \pm 0.020$
$.238 \pm 0.019$
$.331 \pm 0.025$
$.132 \pm 0.023$
$.391 \pm 0.045$

TABLE II Fitting Parameters of the Carreau-Yasuda Viscosity Model for PP and Blends

ning region, λ is a characteristic time at which the Newtonian-pseudoplastic transition occurs, n is the "power law exponent", i.e., the slope of the viscosity curves in the pseudoplastic region, and $\dot{\gamma}$ is the shear rate. Since for polymer neglect, and r is the order of the obtained with $\eta_{\infty} = 0$,²¹ this value was fixed in the calculations, and only the parameters η_0 , a, λ , and n

were estimated using a nonlinear least squares method. The estimated values of these fitting parameters as well as the standard deviations of these values are shown in Table II. For all samples, good fit of the data was obtained with $R^2 > 0.999$ and the standard deviations of the estimated parameter was, in most cases less than 10%.



(a)

(b)



(d)



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Figure 3 (Continued from the previous page)

x2000

Table II shows that all the binary blends presented values of η_0 very similar to that of pure PP. Hence, the order of magnitude of the viscosity of these blends depends only on the matrix characteristics, not being significantly affected by the ultra-high molecular weight PDMS in the SMB_{PP}. The lack of influence of the presence of the ultra-high molecular weight PDMS on η_0 is also observed in the ternary blends, given that both $B_{5(2.81)/20}$ and $B_{5(6.07)/20}$ showed lower values of η_0 than the pure PP and the binary blends. These lower values of η_0 in the ternary blends are due to the presence of the PP-VTES, for the reasons discussed before.

(g)

Before the discussion of the values of the other Carreau–Yasuda viscosity model parameters shown in Table II, it is convenient to analyze the scanning electron micrographs of the surface fracture of pure PP and blends, which are shown in Figure 3. In all blends, the SMB_{PP} phase remained as dispersed domains in the continuous PP matrix. In the binary

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blends [Fig. 3(b,e)], the domain size of the dispersed phase increased with the increase in the SMB_{PP} amount due to the coalescence of the disperse domains. In the ternary blends [Fig. 3(f,g)], the action of PP-VTES in reducing the interfacial energy can be clearly observed since the domain sizes of SMB_{PP} are considerably smaller than in the binary blend B_{0/20} [Fig. 3(e)], which contained the same amount of SMB_{PP}. Indeed, the domain size decreased with the increase in the VTES content of the PP-VTES used.

Another important aspect to be noted about Figure 3 is that in all blends the holes corresponding to the space occupied by the SMB_{PP} presented smooth boundaries, without indication of any appreciable adhesion between the PDMS domains and the PP matrix. Consequently, it can be stated that the PP-VTES molecules act only at the surface level, decreasing interfacial energy around the PDMS domains, but being incapable of generating strong



Figure 4 Storage modulus (G') versus loss modulus (G'') for pure PP (—) and PP/PP-VTES/SMB_{PP} blends at 170°C: 98/0/2 (———), 95/0/5 (——), 90/0/10 (— \bigtriangledown —), 80/0/20 (— \diamondsuit —), 75/5(2.81)/20 (—+—), 75/5(6.07)/20 (—×—).

interaction between these domains and the PP matrix. The behavior of the parameters λ and n of the Carreau–Yasuda model (Table II) in the blends provides support for the latter hypothesis.

Regarding the binary blends, the values of the λ and *n* parameters initially (blend $B_{0/2}$) showed a significant decrease in relation to those of pure PP (by nearly one order of magnitude in the case of the relaxation time λ). Afterwards, these values increased gradually with the increase of the amount of SMB_{PP} in the blend, until reaching values close to those of pure PP for the blend $B_{0/20}$. Decrease of the characteristic time λ means a decrease in the relaxation time of the system and a displacement of the Newtonian-pseudoplastic transition to higher frequencies. The decrease in the relaxation time of the binary blends in relation to the pure PP could be attributed to a very weak interaction and a high interfacial tension between the PDMS and the PP. This combination of factors would lead to an additional relaxation mechanism for the PP matrix, related to the interfacial slip between the uncompatibilized phases. The influence of the nature of the interactions between the phases on the relaxation time of multiphase polymeric systems has already been reported in literature.²²⁻²⁵ On the other hand, the increase in the relaxation time with the increment of the SMB_{PP} content in the binary blends could be related to the increase of dimensions of the dispersed phase [Fig. 3(b,f)], which could lead to a loss of efficiency of the additional relaxation mechanism associated to interfacial slip. The variations in the parameter n could be regarded according to an analogous interpretation since the degree of pseudoplasticity of a material is intimately related to the nature of its relaxation mechanisms.

Even though the interfacial tension of the system has clearly decreased in the ternary blends, as confirmed by the decrease in the dimensions of the dispersed phase [Fig. 3(f,g)], the relaxation times of both ternary blends were lower than that of the pure PP. This fact also indicates that the PP-VTES molecules act only at the surface level, as previously discussed, since an increase in the relaxation time of the system would have occurred if they had generated stronger interaction between the ultra high molecular weight PDMS domains and the PP matrix.^{22,25}

To obtain further information concerning the compatibility of the systems, the storage modulus (G') was plotted against the loss modulus (G'') for the pure PP and for the blends (Fig. 4). It can be seen that the seven curves practically overlap, showing that neither the elasticity nor the rigidity of the PP matrix were significantly changed by the presence of SMB_{PP} and PP-VTES, and confirming that the presence of PP-VTES has affected only the interfacial tension of the system.

DSC studies were carried out to analyze the melting and crystallization behavior of pure polymers and blends in the -65 to 200°C temperature range. Figure 5 shows DSC thermograms for PP, SMB_{PP}, and its blends. Melting (T_m) and crystallization (T_c) temperatures, melting enthalpy (ΔH_m) and degree of crystallinity values for PP and blends are reported in Table I.

As can be seen in Table I, there was practically no change in the melting and crystallization temperatures of the PP matrix when only SMB_{PP} was added. Besides, a slight increase in the degree of crystallinity was observed in these binary blends when the SMB_{PP} content increased. This increase in the degree of crystallinity could be explained on the basis of some degree of nucleation exerted by the SMB_{PP} phase on the PP. Nucleation effect related to SMB domains has also been reported in SMB/HDPE blends.¹²

The ternary blends showed the highest values of degree of crystallinity and also a significant increase in T_c , indicating a more pronounced nucleating effect. Since nucleation is catalyzed by the decrease of thermodynamic barriers,^{26,27} this increase in the nucleating effect of the PP phase of the SMB_{PP} in the ternary blends could be attributed, at least partially, to the decrease of the interfacial energy between SMB_{PP} domains and the continuous PP phase caused by the presence of PP-VTES compatibilizer. This could also explain the increase observed in T_m and ΔH_m when comparing the blends $B_{5(2.81)/20}$ and $B_{5(6.07)/20}$ to the blend $B_{0/20}$. Similar behavior has been observed by Chen and Sun in poly(3-caprolactone) composites.²⁵ The possibility of occurrence of some degree of crosslinking of the PP-VTES compatibilizer, as a result of hydrolysis/condensation reac-



Figure 5 DSC thermograms of melt (a) and crystallization temperature (b) of pure PP (−−−), SMB_{PP} (−−) and PP/PP–VTES/SMB_{PP} blends: 98/0/2 (−−□−), 95/0/5 (−−○−), 90/0/10 (−¬∇−−), 80/0/20 (−¬◊−−), 75/5(2.81)/20 (−+−−), 75/5(6.07)/20 (−−×−−).

tions of ethoxisilane groups of VTES, should also be taken into consideration as another possible cause to the increase of the nucleating effect in the ternary blends. An additional factor contributing to the increase in the crystallinity in the ternary blends is their lower viscosity when compared to the blend without PP-VTES (Fig. 2).

A final important fact to be mentioned is that the removal of the blends from the mixer chamber became easier with the increase of their SMB_{PP} content. This qualitative result shows the effectiveness of the SMB_{PP} as a mold release agent for PP.

CONCLUSIONS

For both binary PP/SMB_{PP} and ternary $PP/SMB_{PP}/PP-VTES$ blends, the morphological evaluation showed

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that the SMB_{PP} remained in the form of dispersed domains in the continuous PP matrix. These domains had their size significantly reduced by the addition of PP-VTES. Rheological and morphological data strongly indicate that PP-VTES acts only by lowering the interfacial tension of the system without generating strong interaction between the PDMS of the SMB_{PP} domains and the PP matrix.

The differences between the blends and the pure PP in terms of relaxation time indicate that the blends presented an additional relaxation mechanism related to the interfacial slip. This hypothesis is in agreement with the fact that the characteristics of the pseudo-plastic behavior of the blends in the melt state were observed to be notably correlated to the dimensions of the SMB_{PP} dispersed phase and the interfacial energy of the system.

The presence of SMB_{PP} domains and the addition of PP-VTES influenced significantly the crystallization and the melting processes of the PP matrix. For both binary and ternary blends, the PP from SMB_{PP} domains showed a nucleating effect leading to an increase in the degree of crystallinity. However, the decrease in interfacial energy and viscosity, promoted by the addition of PP-VTES to the system, led to a more intense nucleating effect and an increase of crystallization, melting temperatures, and melting enthalpy.

The effectiveness of the SMB_{PP} as a mold release agent for PP was observed.

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