SBS and SEBS Block Copolymers as Impact Modifiers for Polypropylene Compounds

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Received 3 February 2004; accepted 23 May 2004
DOI 10.1002/app.21263
Published online in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: Blends of polypropylene (PP) and thermoplastic elastomers (TPE), namely SBS (styrene-butadiene-styrene) and SEBS (styrene-ethylene/1-butene-styrene) block copolymers, were prepared to evaluate the effectiveness of the TPE type as an impact modifier for PP and influence of the concentration of elastomer on the polymer properties. Polypropylene homopolymer (PP-H) and ethylene–propylene random copolymer (PP-R) were evaluated as the PP matrix. Results showed that TPEs had a nucleating effect that caused the PP crystallization temperature to increase, with SBS being more effective than SEBS. Microstructure characterization tests showed that in most cases PP/SEBS blends showed the smallest rubber droplets regardless of the matrix used. It was seen that SEBS is a more effective toughening agent for PP than SBS. At 0°C the Izod impact strength of the PP-H/SEBS 30% b/w blend was twofold higher than the SBS strength, with the PP-R/SEBS 30% b/w blend showing no break. A similar behavior on tensile properties and flexural modulus were observed in both PP/TPE blends. Yield stress and tensile strength decreased and elongation at break increased by expanding the dispersed elastomeric phase in the PP matrix.© 2004 Wiley Periodicals, Inc.

Key words: poly(propylene); block copolymers; blends; mechanical properties; morphology

INTRODUCTION

Poly(propylene) (PP) is one of the most widely used polymers, seen in the form of molded pieces, sheets, films, and fibers. This versatility is a result of its properties, which allow it to be processed in several different ways. In some special applications, PP blends using other polymers have been used successfully, mainly in applications requiring high fracture toughness. These blends are used to create new materials with synergic properties. The development of the multiphase morphology during the mixture of the molten polymers plays an important role in the blend properties.1 One of the most successful outcomes of polymer blends is the improvement in impact strength of PP at low temperatures, generally achieved by incorporating an elastomer into the PP matrix.2–4 Chemical modifications of the blend components and their compatibilization agent have been used to improve rubber dispersion in the PP matrix as well the mechanical properties of the blend. The compatibility and interfacial adhesion between the dispersed elastomer particles and PP matrix5 have been correlated with the impact strength of the blend. It has been shown that the mechanical properties of multiphase polymeric materials are strongly affected by the morphology of the dispersed phase.

The incorporation of a rubbery phase in PP can be achieved by the copolymerization of ethylene–propylene in the polymerization process6 or by mechanical blending with EPDM7–9 or EPR,10,11 usually employed as impact modifiers for PP. Ethylene-co-but-1-ene rubber (EBR)3 showed higher toughening efficiency when compared with EPR. More recently, poly(ethylene-co-octene) was proposed as an impact modifier for PP, showing good toughening efficiency12 and better processability13 compared to EPDM. However, thermoplastic elastomers (TPE) like SBS polystyrene-block-polybutadiene-block-polystyrene or SEBS polystyrene-block-poly(ethylene-co-but-1-ene)-block-polystyrene14,15 have higher service temperature and better solvent resistance than the classical butadiene-based rubbers and metalocenic poly(ethylene-co-α-olefins). Thus, PP blends with SBS and SEBS hold the promise of improved properties in relation to those obtained using conventional elastomers and this widens the scope of applications of these materials. SBS and/or SIS (polystyrene-block-polysoprene-block-polystyrene) were used as a compatibilizing agent in PP/PS blends16,17 to obtain a well-dispersed phase morphology. PP/PS blends with these elastomers showed lower poly(styrene) domains size, enhancing
toughness and impact strength, than the ones not compatibilized. SEBS also was used as a compatibilizing agent in PP/LLDPE blends, improving the adhesion by lowering the interfacial tension between the components. Maleinated SEBS (SEBS-g-MA) is also used as a compatibilizing agent in polypropylene/polyamide blends prepared by reactive blending.

Isotatic polypropylene (i-PP) and SBS blends produced by mechanical blending show good tensile properties and impact strength, which can be improved further by dynamic vulcanization. Maleinated SBS (SBS-g-MA) were used advantageously compared to SBS as impact modifiers for PP due to the crosslinking of SBS-g-MA and to the formation of a graft copolymer between i-PP and SBS-g-MA. Several previous studies have shown that the impact resistance of i-PP increased significantly and its yield modulus and stress decreased when SEBS was added. Blends of syndiotatic polypropylene (s-PP) with SEBS were also evaluated and it was seen that SEBS has a good interfacial adhesion with both i-PP and s-PP surfaces and produces similar changes on tensile and impact behavior.

The aim of the present study is to evaluate simultaneously the effectiveness of the TPE type as an impact modifier and the influence of the PP matrix and the TPE concentration on the thermal and mechanical properties of the PP/TPE blends. Two polypropylene varieties (a PP homopolymer and a PP copolymer) and two thermoplastic elastomers (SBS and SEBS) were evaluated and any differences found in their properties were correlated with the blend morphology.

**EXPERIMENTAL**

**Preparation of materials and blends**

The polypropylenes used in the blends were commercial grade polypropylene homopolymer (PP-H) and poly(propylene-ran-ethylene) copolymer (PP-R) manufactured by Braskem S.A. (Triunfo/RS, Brazil). The thermoplastic elastomers were commercial grade block copolymers SBS and SEBS supplied by Kraton S.A. All samples were used as supplied.

The components of the PP/TPE blends were physically mixed and then extruded in an Oryzon single-screw type extruder (diameter 25 mm; length = 36) operated at 60 rpm in a barrel melt mixing temperature profile of 200/220/220/230/230/200°C, where the latter refers to the die temperature. The elastomer concentrations in the blend were 5, 10, 15, 20, and 30% b/w. The PP/TPE blends were prepared using the same set of mixing conditions. The PP and PP/TPE blends test specimens (TS) were injection-molded in a Battenfeld PLUS 350 injection molder according to ASTM D 4101. The TSs dimensions corresponded to those specified in ASTM D 638 and ASTM D 790.

**Thermal characterization**

The thermal behavior of the PP/TPE blends and the starting polymers was analyzed in a TA Differential Scanning Calorimeter (DSC) model 2010. TSs were heated to 200°C, kept at this temperature for 5 min, and then cooled to room temperature at 10°C/min. They were then reheated to 200°C at the same heating rate. Crystallization and melting temperatures (T_c and T_m) and fusion enthalpy (ΔH_m) were taken from the second and third run curves, respectively. Sample crystallinity content was calculated using a PP fusion enthalpy reference value of 190 J/g.

**Scanning electron microscopy**

Blend morphology was evaluated in a Jeol JSM 5800 Scanning Electron Microscope (SEM). The specimens were cryogenically fractured after 2 min in liquid nitrogen and the TPE or rubber particles in the fractured surface were etched with tetrahydrofuran (THF) at 60°C for 30 min. Samples were dried and then sputter-coated with gold in a sputter coater. SEM analysis was carried out on the cryogenic fracture surfaces using an acceleration voltage of 20 kV. The mean particle size of the TPE droplets domains for PP/TPE blends was determined with a Leica software image analyzer.

**Mechanical characterization**

The tensile properties of the PP and PP/TPE blends were evaluated in an Instron Tensile Machine model 4202 according to ASTM D 638–95 using a crosshead speed of 2 mm/min. The flexural modulus was determined using an Instron Tensile Machine model 4466 according to ASTM D 790–95a. The Izod impact strength of notched specimens was determined in a CEAST model 6845–800 Impact Machine according to ASTM D 256.

**RESULTS AND DISCUSSION**

The PP-H and PP-R varieties used in this work show differences in their structure and chemical composition. The former is a highly isotatic variety with higher stiffness than PP-R. Conversely, PP-R is a propylene-ethylene random copolymer with lower crystallinity and better impact resistance. SBS and SEBS are linear triblock copolymers with approximately 30% styrene b/w in their composition and a different middle block polymer, namely polybutadiene and poly(ethylene-co-but-1-ene), respectively.

Table I shows PP and TPE melt index, complex viscosity (η*) at 10 Hz, molecular weight (M_w) and polydispersity (M_w/M_n). The M_w and M_w/M_n of the PP samples is of the same magnitude as the TPE samples. A small difference between the molecular
weights of the polymer pairs, PP or TPE, was intentionally selected to minimize the effect of $M_w$ on the properties of the blends. The $M_w$ and polydispersity of the PP samples correspond to approximately 240,000 g/mol and 4.0, respectively. The $M_w$ of the TPE samples is approximately 80,000 g/mol and their polydispersity is lower than 2.0 due to the anionic mechanism of polymer synthesis.

PP and TPE show marked differences in complex viscosity. In addition, the $\eta^{*}_{TPE}/\eta^{*}_{PP}$ ratios at 10 Hz were of the same magnitude, with the SEBS/PP viscosity ratio (approximately 55) being about 25% higher than the SBS/PP ratio (approximately 39). Figure 1 shows the changes in complex viscosity of PP and TPE versus frequency. It can be seen that the polymer pairs present the same curve profiles. The complex viscosity curves of TPE show a more dramatic decrease than those of PP. The effect of viscosity ratio on blend morphology is widely known. Therefore, it is expected that blends of PP/TPE should present a similar viscoelastic behavior when the melts are mixed and any difference in the morphology of the blends must be the sole result of the chemical nature of the components. The triblock copolymers SBS and SEBS have a different midblock, a polybutadiene (PB) and an ethylene-but-1-ene (EB), respectively. Therefore, the current study analyzed the ability of a styrene thermoplastic elastomer, particularly in terms of its chemical and physical nature, to change the thermal and mechanical properties of PP whose matrix was more or less crystalline, as well as the morphology of the resulting blend.

**PP/TPE thermal behavior**

Figure 2 compares the fusion peaks of the endothermic DSC curves for PP-H and PP-H/SBS 10% b/w (−−−−) and SEBS 10% b/w (—) blends.

**Figure 1** Complex viscosity of PP-H, PP-R, SBS, and SEBS at 200°C.

**Figure 2** Fusion peak of endothermic DSC curve of PP-H and PP-H/SBS 10% b/w (−−−−) and SEBS 10% b/w (—) blends.

### Table I

<table>
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<tr>
<th></th>
<th>Polypropylene</th>
<th>TPEs</th>
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<tbody>
<tr>
<td></td>
<td>PP-H</td>
<td>PP-R</td>
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<tr>
<td>MI (g/10min)</td>
<td>11.6$^a$</td>
<td>10.7$^a$</td>
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<tr>
<td>$\eta^*$ (10 Hz)</td>
<td>632</td>
<td>660</td>
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<tr>
<td>$\eta_{TPE}/\eta_{PP}$</td>
<td>40$^e$/56$^d$</td>
<td>38$^e$/53$^d$</td>
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<td>$M_w$</td>
<td>232,600</td>
<td>250,800</td>
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<td>568,400</td>
<td>615,400</td>
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<td>$M_w/M_x$</td>
<td>4.3</td>
<td>4</td>
</tr>
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</table>

$^a$ 230°C and 2.16 kg.

$^b$ 200°C and 5 kg.

$^c$ SBS/PP.

$^d$ SEBS/PP.

$^e$ 230°C and 2.16 kg.

$^f$ 200°C and 5 kg.

$^g$ SBS/PP.

$^h$ SEBS/PP.
or 10% SEBS b/w. The addition of TPE to PP-H did not affect its melting temperature ($T_m$), taken as the apex of the endothermic peak. However, the width of endothermic peak was reduced. A similar behavior was observed in all PP/TPE pairs but the peak width was dependent on the TPE content.

Figures 3 and 4 show a comparison of the crystallization peaks of exothermic DSC curves of pure PP-H and PP-R and their blends, respectively. For both PP varieties, regardless of TPE type, the crystallization peak was displaced to higher temperatures in relation to unmodified PP. This demonstrates that TPE acts as a nucleation agent for PP. The highest curve displacement and crystallization temperature ($T_c$), measured as the apex of the crystallization peak, was found in the PP/TPE 5% b/w blends. PP blends with higher TPE contents have an exothermic peak falling between that of pure PP and that of the 5% b/w blend. All crystallization peaks of the PP/TPE blends were slightly narrower than those of pure PP, so the TPE growth nuclei predominate over all the spontaneous PP nuclei. It was also seen that SBS was more effective as a nucleation agent than SEBS for both PP-H and PP-R due to the higher crystallization temperature ($T_c$) shown by the PP/SBS blends.

The results showed that SBS displaces the crystallization temperature of PP blends to higher values than SEBS does. Ferrer et al. and Saroop and Mathur did not report any changes in the crystallization peak width and crystallization temperature in their studies with isotactic-PP/SBS blends. However, the PP samples in their research had a higher $M_w$ than the PP used in the current study, with higher viscosity and consequently lower viscosity ratio. In their studies, other preferential interactions such as the interdiffusion of macromolecular segments were probably present.

Table II shows the $T_c$, $T_m$, $\Delta H_m$ and crystallinity values for all PP/TPEs blends, where the PP-H and
PP-R melting temperatures correspond to 163 and 142°C, respectively. The latter is lower than the former due to the ethylene insertion in the polypropylene chain, which disrupts the isotactic sequences, decreases the size of lamellae and introduces defects in the crystallite. The SBS and SEBS additions in both the PP homopolymer and the random copolymer caused their fusion enthalpy to change and this indicates that they affect polymer crystallinity. For PP-H blends there was an increase in $\Delta H_m$ of up to 10% b/w of the TPE content. For higher TPE amounts, $\Delta H_m$ showed no change with SBS but a decrease was observed when SEBS was used. On the other hand, for PP-R blends, an increase in $\Delta H_m$ was seen regardless of the TPE type. Nevertheless, Wilhem and Felisberti reported in their studies by DSC and X-rays that SBS and SBS-g-MA did not affect i-PP crystallinity in i-PP/SBS and i-PP/SBS-g-MA blends.

The influence of TPE as a nucleation agent can be related to its styrene blocks ($T_g \approx 85^\circ$C), which have more rigid or inflexible segments than polypropylene ($T_g \approx -15^\circ$C). Thus, the PS domains function as nuclei and induce the PP macromolecule segments to organize as a three-dimensional unit at a higher temperature than usual. The PS block of SBS or SEBS produced a similar change in the PP crystallization temperature, considering that in the PP crystallization temperature range (105 to 125°C) the PS block is very close to its glass temperature region (between 80 to 100°C) and is much less flexible than the PP segments. As TPEs have the same styrene content (approximately 30% b/w) and the SBS has a stronger influence as a nucleation agent, the PB midblock should have more influence on the performance of the PS crystallization nuclei. The polyolefin block or the EB segment of the SEBS has a higher affinity or compatibility with the PP macromolecules. In the melt state this probably causes the styrene block to disperse better into the PP matrix, in this way affecting the PS segments or their mobility much more compared with the PS blocks in the SBS. Setz in his study of i-PP/SBS blends stated that EB segments move into i-PP macromolecules and aggregate to form micelles. Therefore, the SEBS could hinder the access of PP segments to the growth nucleus, resulting in a less crystalline matrix. The PB block or segment in SBS does not show a good affinity with the PP macromolecule so the PS segments are less affected than those in SEBS. For this reason, they have less mobility and act more efficiently as a nucleation agent. The matrix crystallinity results listed in Table II show that both TPEs caused the matrix crystallinity to increase. This contributes to an improved mechanical response considering that the addition of elastomers to semicrystalline polymers always reduces the flexural modulus of the blend.

### PP/TPE microstructure morphology

The formation and breakup of droplets or domains in viscoelastic fluids depend on the molecular weight and viscosity of the components, the viscosity ratio, the interfacial tension, the flow type, and the blend composition. Like other rubber-modified polymers, PP/TPE blends separate into distinct phases. The PP matrix and TPE type influence the rubber droplet size, shape, and distribution, resulting in blends with different morphologies, particularly for higher TPE contents. If better blend compatibility is to be achieved, it is crucial to promote good interaction at the interface of the components so that a stabilization of the melt morphology during melt processing is obtained.

Table II shows the SEM micrographs of blends of PP/TPE 30% b/w. The SEBS droplets or domains [Figs. 5(a) and (c)] were smaller than those of SBS in both PP matrices, with low droplet coalescence levels.
The coalescence of several small droplets resulted in larger rubbery domains being formed in the PP/SBS blends, as can be seen in the several unevenly shaped holes in Figures 5(b) and (d). Wilhem and Felisberti\(^\text{35}\) also reported an increase in the elastomer average particle size in PP/SBS blends for higher SBS content, attributed to elastomer particles coalescence.

Table III presents the mean diameter values of rubber droplets in relation to the TPE type used and its concentration in the blend. Coalescent domains [like those in Figs. 5(b) and (d)] exceeding 4 \(\mu\)m were excluded from the mean diameter calculation. The rubber particle size of all PP/TPE blends was between 1.0 and 2.5 \(\mu\)m while the average diameter of SBS rubber particles was higher than that of SEBS particles.

The mean diameter of rubber particles can be related to a more or less effective breakup of the TPE into the PP matrix. Even when the SEBS content in the blend was increased, the average particle size did not change significantly. Since the viscosity ratios \(\eta_{TPE}/\eta_{PP}\) were slightly higher for SEBS than SBS, in the case of the former, PP has a higher ability to transfer shear stress to the rubber phase and break it apart. The PP-H/SBS blends with higher rubber content had an average particle diameter (2.4 \(\mu\)m) that was nearly twice as big as that of SEBS blends. However, for a less crystalline matrix, the higher SBS content (PP-R/SBS 30% b/w) decreases the average particle diameter and becomes similar to the particle size found in SEBS. This demonstrates that the chemical nature of the

![Figure 5: SEM image of PP/TPE 30% b/w blends (×3,000). (a) PP-R/SEBS; (b) PP-R/SBS; (c) PP-H/SEBS, and (d) PP-H/SBS.](image)

### TABLE III

<table>
<thead>
<tr>
<th>TPE (wt %)</th>
<th>PP-H</th>
<th>PP-R</th>
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<tr>
<td></td>
<td>SEBS</td>
<td>SBS</td>
</tr>
<tr>
<td>10</td>
<td>1.14 ± 0.7</td>
<td>1.54 ± 0.7</td>
</tr>
<tr>
<td>20</td>
<td>2.51 ± 0.8</td>
<td>2.37 ± 0.8</td>
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<tr>
<td>30</td>
<td>1.59 ± 0.4</td>
<td>2.34 ± 0.8</td>
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* Standard error less than 0.2 \(\mu\)m.
matrix also influences the TPE breakup into the PP. As mentioned before, the EB blocks in the SEBS have more affinity with PP macromolecules, thus they better disperse the TPE in the melt state. For the SEBS content used in this work, very low particle coalescence was observed, as already pointed out by Stricker et al., who observed no coalescence level and smaller SEBS domains in blends with PP.

**PP/TPE mechanical properties**

The blend morphology reflects directly on the impact strength of the material and any changes in microstructure cause properties to change. For this reason, the blend crystallinity is also an important factor. A suitable morphology with smaller, well-dispersed rubber domains is desirable to yield high polymer toughness. Table IV displays the values of PP/TPEs Izod impact strength at 0° and 23°C, yield stress, and elongation at break as a function of the PP matrix and TPE content.

Low TPE concentrations such as 5% b/w resulted in low impact behavior changes of the blend, with a gradual increase of this to 30% b/w appearing to be far more effective. No break response at 23°C was observed for the PP-R/TPE 30% b/w blends. This can be attributed not only to the amount of rubber present but also to its morphology or rubber particle size (around 1.5 μm). This is also corroborated by the no break response of the PP-R/SEBS 30% b/w, a more crystalline matrix that has a similar mean particle diameter. On the other hand, the PP-H/SEBS 30% b/w with around 2.4 μm domains showed an Izod impact energy of 252 J/m at 23°C. This value is seven times higher than the 35 J/m observed for pure PP-H. The PP-R/SEBS 30% b/w did not break even at 0°C, illustrating the influence of the PP matrix on the impact strength. The PP-R/TPE blends presented a slightly higher impact strength than the PP-H blends in all compositions with over 10% TPE b/w. When SEBS was blended with the PP random matrix, the smallest and best dispersed TPE domains were obtained, resulting in the material with the best impact response at both temperatures.

The impact strength results may be also a consequence of the interactions on the interface between the matrix and the elastomer. SEBS has a better interaction with the PP matrix than SBS due to its EB midblocks being more compatible with the noncrystalline PP fraction, which facilitates segmental diffusion between the matrix and the rubber particles on the interfaces. In addition, PP-R contains a fraction of highly modified ethylene that favors the interaction between TPE particles and the PP matrix.

Differences in the values for tensile properties in the PP/TPE blends were observed due to the matrix type and TPE content. The PP/TPEs yield stress and elongation at break values shown in Table IV were taken from stress–strain curves, such as the PP/SEBS blends curves shown in Figure 6. The reduction in yield strength for increased TPE content was similar for all PP/TPE blends, and the TPE type had no effect. According to Stricker et al. and Gupta and Purwar, differences in morphology did not bring changes on yield stress. However, the elongation at break of PP/TPE blends presented large differences thanks to the type of PP matrix and elastomer in all blend compositions investigated. The PP/SEBS blends showed greater deformation and did not break even at over

<table>
<thead>
<tr>
<th>TPE (wt %)</th>
<th>Yield Stress (MPa)</th>
<th>Elongation at break (%)</th>
<th>Impact strength (J/m) 23°C</th>
<th>Impact strength (J/m) 0°C</th>
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<tr>
<td></td>
<td>SBS</td>
<td>SEBS</td>
<td>SBS</td>
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a Notched Izod; nd, not determined; nb, no break.
800% elongation. PP-R, due to its less crystalline nature, elongated 440% while PP-H elongated 180%.

The yield stress of PP-H/SEBS blends falls across the entire composition range evaluated while the yield peak widens due to the more elastomeric nature of the blend [Fig. 6(a)]. These results are in agreement with published studies, in which PP with SBS and SEBS blends showed decreased yield stress, tensile strength, and tensile modulus and increased elongation at break.

The elongation at break increased gradually with the addition of SBS, but for PP-H/SBS 10% b/w it was lower than for pure PP-H, showing that the blend has a lower ability to withstand deformation under stress. This behavior could be related to the higher matrix crystallinity or an overcrystallization of PP caused by orientation under stress in the plastic deformation region. On the other hand, the PP-H/SEBS 30% b/w had a stress–strain curve similar to PP-R/SEBS blends [Fig. 6(b)].

Despite the strong decrease in the yield stress, the stiffness of PP/TPE blends did not follow the same pattern. The addition of rubber to a semicrystalline polymer usually improves its impact resistance; however, a nondesirable side effect is the reduction in stiffness. If TPE is used as impact modifier, some degree of reduction in yield stress can be tolerated in favor of a large improvement in impact resistance. As TPEs are stiffer than conventional elastomers, they may result in better performance blends. This occurs because of their two-phase morphology, where the SBS polystyrene particles are dispersed in the polybutadiene matrix. Since the PS glass transition temperature is approximately 100°C, and both SBS and SEBS contain approximately 30% PS b/w, they are classified as a stiff material. The stiffness of the PP/TPE blends is assessed in terms of their flexural modulus. The variation of the PP/TPE flexural modulus in relation to their SBS and SEBS content is shown in Figure 7.
As expected, the PP-R blends displayed lower stiffness than PP-H blends and their flexural modulus decreased as concentrations of TPE increased. In addition to TPE content, the matrix type also affected the flexural modulus. The chemical difference between the TPEs did not affect blend stiffness as much as reported by other authors.26 The flexural modulus curves of PP-R blends showed a smaller angular coefficient, with a lower variation in stiffness than the PP-H blends with a more crystalline matrix.

To sum up the findings on morphology and mechanical properties, the mechanical response of PP was affected by the addition of TPE and the SEBS was shown to be a better impact modifier for both PP matrices with acceptable loss in the modulus values.

CONCLUSIONS

Both SBS and SEBS behaved as a nucleation agent in both isotatic polypropylene (PP-H) and polypropylene random copolymer (PP-R). SBS had a more dramatic effect compared to SEBS in both PP matrices, resulting in higher $T_c$ values. Although the nucleation effect may change the crystallinity of the matrix in the blends, it seems to affect impact strength much less compared with the rubber dispersion or blend morphology.

The average diameter of rubber particles and their dispersion in the PP/SEBS were lower and better distributed than in the PP/SBS blends, resulting in a material with improved impact strength and excellent Izod impact resistance at 0 and 23°C due to a more appropriate morphology.

PP/TPE blends with both elastomer types showed a similar reduction in tensile properties such as yield strength and flexural modulus. However, the elongation at break was more significant for the PP-R/SEBS blend, which exhibited a more elastomeric nature. Nevertheless, both elastomers can be used as impact modifiers, resulting in the same degree of modification in tensile properties.

The authors thank CNPq for the financial support to this study. In addition, they are indebted to Braskem S.A. for the materials and analysis.

References