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From Disposal to Technological Potential: Reuse of Polypropylene Waste from Industrial Containers as a Polystyrene Impact Modifier

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Abstract: The practice of recycling over the years has been increasingly encouraged, with the aim being the manufacturing of materials that contribute to sustainable development. In light of this, the present work evaluated the potential of mixtures of polystyrene (PS)/recycled copolymer polypropylene (PPr), using styrene-(ethylene/butylene)-styrene (SEBS) as a compatibilizing agent. Initially, the mixtures were prepared in a co-rotational twin-screw extruder, and, afterwards, the extruded granules were molded by injection. The properties of torque rheometry, impact strength, tensile properties, differential scanning calorimetry (DSC), heat deflection temperature (HDT), and scanning electron microscopy (SEM) were evaluated. The formulation PS/PPr/SEBS (70/20/10 %wt.) demonstrated an increase in viscosity, corroborating with an increase of 123% and 227% in the elongation at break and impact strength, respectively, compared to neat PS. Though the elastic modulus and tensile strength suffered losses, the reduction was not drastic. Furthermore, the addition of a semi-crystalline recycled material in the amorphous matrix (PS) contributed to an increase in thermomechanical strength, as seen in the HDT. The morphology revealed that SEBS is effective in making PS/PPr mixtures compatible because the dispersed phase is well adhered to the PS matrix and promotes greater morphological stability. Thus, it is possible to add value to discarded material and reduce the costs of the final product, which can reduce pollution.

Keywords: sustainable development; polypropylene; reuse; impact modifier; polystyrene

1. Introduction

As the world's population grows, an enormous amount of waste is generated that is derived from polymeric materials, contributing to environmental and social impacts [1]. Polypropylene (PP) is one of the most requested polymers in the world for developing manufactured goods such as food packaging, bags for grains and fertilizers, plastic chairs, industrial containers, toys, and plastic cups [2,3]. High consumption of PP leads to an increased production of post-industrial waste and, consequently, the practice of mechanical recycling is encouraged due to PP's beneficial properties [4].

PP is a semicrystalline polymer known commercially for presenting an excellent balance of properties, including high chemical resistance, reasonable ductility, good stiffness, low density, and excellent processability [5,6]. Therefore, developing polymeric mixtures with recycled PP is



increasingly explored, because it allows the reduction of waste deposited in the environment and contributes to the production of materials with good properties [7].

The heterophasic copolymer polypropylene (PP) is a complex material. It has both a homopolymer PP matrix and dispersed ethylene/propylene particles. Generally, copolymer PP is used in applications that require high impact strength, such as industrial containers [8]. At present, Brazil is one of the largest PP producers worldwide, and it has the largest industrial park in Latin America for the production of industrial containers [9]. This stems from the high demand for industrial containers. In this context, the production of post-consumption waste is high, and waste also arises from the need to dispose of damaged industrial containers derived from copolymer PP. Copolymer PP residues are a source of environmental pollution and raw material waste. They are also associated with beneficial properties and high added value, including the technological potential to be used as a polystyrene (PS) impact modifier.

As a transparent and fragile thermoplastic with good tensile properties, PS can easily be synthesized, processed, and recycled, and it is resistant to biological degradation [10]. However, it has low impact strength and, therefore, it must be toughened for certain applications [11]. PS toughening has already been explored in the literature with styrene-butadiene (SBR), styrene-butadiene-styrene (SBS), styrene-butylene/ethylene-styrene (SEBS), and ethylene propylene-diene (EPDM) rubbers [12–16]. However, despite the fact that these rubbers achieve satisfactory PS impact strength, they severely reduce tensile properties and thermomechanical strength. To improve the properties of polystyrene without incurring severe losses in tensile properties, the research [17–19] was directed towards polystyrene/polypropylene mixtures using compatibilization agents.

Samsudin et al. [20] investigated the chemical resistance of polystyrene (PS)/polypropylene (PP) mixtures compatibilized with SEBS. The results showed that the chemical resistance of PS to acetone and tetrahydrofuran (THF) increased. The chemical resistance of PS/PP mixtures increased with the increase in the SEBS content and a relatively low PP content. However, it decreased when the SEBS content increased and a high PP content was used. Semicrystalline polypropylene has been shown to increase the chemical resistance of PS.

Halimatudahliana et al. [21] evaluated the effect of several compatibilizers on the mechanical properties of polystyrene/polypropylene mixtures. The PS/PP blends in the compositions 20/80, 50/50, and 80/20 (% by weight) were prepared in a single screw extruder. In general, styrene-(ethylene/butylene)-styrene (SEBS) was the best compatibilizer for PS/PP mixtures. When the PS/PP mixture was made compatible with 7.5%, toughness was improved in all compositions. Additionally, the fragile behavior of polystyrene was converted to extreme ductility with PS/PP mixtures compatible with SEBS.

Latreche et al. [22] developed polystyrene/polypropylene mixtures compatibilized with several compatibilizers. Generally, the compatibilizer tended to reduce the interfacial tension between the phases, generating greater morphological stability, in comparison with the non-compatibilized blend. The ductility and impact strength properties were enhanced when the PS/PP blend was compatibilized with SEBS. However, there was a reduction in microhardness and in the degree of crystallinity, indicating an increase in the flexibility of the PS/PP/SEBS blends.

Luna et al. [23] investigated the toughening of polystyrene (PS) with hybrid mixtures of copolymer polypropylene (PP) and recycled polypropylene (PPr), using SEBS as a compatibilizing agent. The results showed that the mixtures presented a pseudoplastic fluid behavior, according to complex viscosity analysis. There was an increase in impact strength, elongation at break, thermal stability (TG), and heat deflection temperature (HDT) of the mixtures, compared to neat PS. For PS/PP/PPr/SEBS mixtures in the investigated composition range of PPr (7%, 14%, and 21% by weight), there were no significant differences in the properties of the mixtures. In this case, a greater amount of recycled material can be used without compromising properties.

The mixture of polystyrene (PS) and polypropylene (PP) is one of the most studied, mainly due to the low cost and technological relevance of these materials [24–26]. Most PS/PP mixtures are developed

using homopolymer PP and, at the same time, it is not recycled. Despite PP's potential to improve the properties of polystyrene, there is practically no work in the literature that used recycled copolymer PP to toughen PS. Both the economic and environmental benefits are justification for research evaluating the potential of this recycled material.

This work aimed to study polystyrene/recycled copolymer polypropylene mixtures, using styrene-(ethylene/butylene)-styrene (SEBS) as a compatibilizing agent.

2. Experimental

2.1. Materials

The crystal polystyrene (PS), marketed under code U249, density of 1.04 g/cm³ and melt flow index (MFI) of 19 g/10 min (200 °C/5 kg), was supplied in the form of pellets and manufactured by Unigel SA (Brazil).

The heterophasic copolymer polypropylene (called PPv; "v" refers to virgin material), sold under the EP440P code, in the form of pellets, density of 0.895 g/cm³ and MFI of 17 g/10 min (230 °C/2.16 kg), was purchased from Braskem. This copolymer has ethylene-propylene (EP) dispersed in the polypropylene matrix.

The recycled copolymer polypropylene (called PPr; "r" refers to recycled material), with a density of 0.904 g/cm3 (ASTM D792) and MFI of 36 g/10 min (230 °C/2.16 kg) was from industrial containers. The mechanical, thermomechanical, thermal, rheological properties, and morphology of this recycled material can be accessed in the literature [27].

The styrene-(ethylene/butylene)-styrene (SEBS) copolymer that was used as a compatibilizer agent, in powder form, under code G1652, containing 30% styrene and MFI of 5 g/10 min (230 $^{\circ}$ C/5 kg), was supplied by Kraton.

2.2. Methods

2.2.1. Preparation of Recycled Polypropylene (PP_r)

The containers were crushed in a knife mill, giving rise to the flakes. Subsequently, the flakes were washed with distilled water to remove contaminants and subsequently dried in a vacuum for 24 h at a temperature of 60 °C. Figure 1 shows a schematic representation of the obtain the recycled material.



Figure 1. Schematic representation of the method used to obtain recycled material.

2.2.2. Blends Preparation

The blends were dry-mixed and subsequently processed in a modular co-rotational twin screw extruder (model ZSK (D = 18 mm and L/D = 40) from Coperion Werner & Pfleiderer) at 200 °C in all zones, a screw rotation speed of 200 rpm, and a feed rate of 3 kg/h, with the screw profile configured with distributive and dispersive mix elements. Subsequently, the materials were granulated and dried in a vacuum oven for 24 h at 60 °C. Polystyrene (PS) was processed and dried under the same conditions as the mixtures. Table 1 illustrates the weight proportions (%) of the compositions that were used in the development of the mixtures.

Table 1. Mixture compositions with weight proportions (%). PS: polystyrene; SEBS: styrene-(ethylene/butylene)-styrene; PPv: copolymer polypropylene; PPr: recycled polypropylene.

Sample	Proportion (%wt.)		
PS	100		
PS/SEBS	95/5		
PS/SEBS	90/10		
PS/PPv	70/30		
PS/PPr	70/30		
PS/PPv/SEBS	70/25/5		
PS/PPr/SEBS	70/25/5		
PS/PPv/SEBS	70/20/10		
PS/PPr/SEBS	70/20/10		

2.2.3. Injection Molding

The materials were injection molded in an Arburg injector (Model Allrounder 207C Golden Edition) for obtaining impact, traction, and HDT specimens, according to the ASTM D256, ASTM D638, and ASTM D648 standards, respectively. The injector contains five heating zones, with a temperature profile of 190 °C being adopted for all zones. The molding conditions were: mold temperature of 20 °C; cooling time inside the mold of 25 s; and injection and discharge pressure of 800 and 500 bar, respectively. Figure 2 shows a schematic flowchart of the processing performed in the laboratory.

Characterizations

Rheological curves were obtained in a Thermo Scientific Haake PolyLab QC mixer, with roller-type rotors at 200 °C, with a rotor speed of 60 rpm, under air atmosphere for 10 min.

An Izod impact strength test was performed on the notched specimens, according to ASTM D256, in a Ceast model Resil 5.5 J, operating with a hammer of 2.75 J, at room temperature (~23 °C). Results were analyzed with an average of seven specimens.

A tensile test was performed on the injected specimens, according to ASTM D638, using an universal test machine, EMIC DL 2000, with a rate of 50 mm/min and load cell of 20 kN, at room temperature (~23 $^{\circ}$ C). Results were analyzed with an average of seven specimens.

Heat deflection temperature (HDT) was measured according to ASTM D648 in a Ceast model HDT 6 VICAT, with a voltage of 455 kPa and a heating rate of 120 °C/h (method A). The temperature was determined after the sample deflected 0.25 mm. Results were analyzed with an average of three specimens.

Differential scanning calorimetry (DSC) analysis was performed in a DSC-Q20 from TA Instruments. The scans were computed from 30 to 180 °C, under a heating rate of 10 °C/min, gas flow rate of 50 mL/min, in nitrogen atmosphere; samples of approximately 7 mg were tested.

Scanning electronic microscope (SEM) analysis was obtained with a Shimadzu SSX-550 Superscan device with a 30 kV voltage under high vacuum conditions. The fracture surfaces of impact samples were analyzed. The surfaces were coated with a gold layer.



Figure 2. Schematic representation of PS processing, binary mixtures (PS/PPv and PS/PPr), and compatibilized mixtures.

• Statistical Analysis

The statistical analysis was performed using the Microsoft Excel program. The arithmetic mean (\overline{X}) represents a measure of centrality, the estimate of which is calculated by:

$$\overline{X} = \frac{1}{n} \sum_{i=1}^{n} x_i, \tag{1}$$

where *n* represents the number of samples and x_i represents the property value.

The standard deviation is a measure of dispersion, that is, it indicates how uniform the data set is. The deviation can be estimated by:

$$\delta^2 = \frac{1}{n} \sum_{i=1}^n \left(x_i - \overline{X} \right)^2,\tag{2}$$

where *n* represents the number of samples and x_i represents the property value; \overline{X} is the arithmetic mean of the property.

3. Results and Discussion

3.1. Torque Rheometry

Torque rheometry is a technique that provides evidence of degradation, crosslinking, chemical reactions, processability, and increased compatibility of polymeric mixtures [28]. Figure 3 shows the torque curves as a function of the processing time for PS, SEBS, PPv, and PPr. SEBS presented the highest torque (~13.5 N.m) at the end of processing and, consequently, the highest viscosity when compared to other neat polymers. Such behavior indicates a low flow of SEBS, generating greater processing difficulty. PS has a higher viscosity compared to PPv and PPr, due to the presence of styrenic groups, which decrease the flow. Apparently, PPv should have a higher viscosity, because it has a higher MFI (17 g/10 min) compared to neat PS (19 g/10 min). This inversion of change in viscosity between PS and PPv is related to the different processing conditions in the MFI test (temperature and load). PPv showed a higher viscosity than PPr, suggesting a higher molecular weight and, as a result, higher torque. The PPr, being a recycled material, undergoes chain splitting with each reprocessing, causing a reduction in molecular weight and viscosity, as reported in the literature [29].



Figure 3. Torque curves as a function of the processing time of neat polymers.

Figure 4 illustrates the torque curves as a function of the processing time for the PS, the binary mixtures (PS/PPv and PS/PPr), and the compatibilized mixtures.

Figure 4 shows that, after 5 min of the process, the torque tends to be practically constant for all materials, suggesting there is no occurrence of degradation. This behavior indicates viscosity stability for the process conditions used. Theoretically, this is the working viscosity of the material; i.e., the point to be extruded or injected [30]. Polystyrene (PS) showed higher torque when compared to the binary blends PS/PPv and PS/PPr and, therefore, higher viscosity. The low viscosity of the binary blends indicates that there is no interactions between PP and PS due to structural differences, generating an incompatibility in the system [31].

The PS/PPv/SEBS and PS/PPr/SEBS blends, regardless of SEBS content, increased viscosity compared to the PS and binary blends, suggesting an increase in compatibility. Apparently, the viscosity of the PS/PPv/SEBS and PS/PPr/SEBS blends was strongly influenced by the SEBS content. The torque value of the compatibilized blends was intermediate between the PS and SEBS, with an increase in torque with an addition of SEBS content. Probably, as SEBS has a low melt flow index compared to

PS and PP, it contributed to an increase in the viscosity and torque of the blends. However, there is evidence of interactions between similar chemical groups, such as the SEBS styrene blocks with PS, as well as ethylene/butylene (EB) with PP, favoring the increase in viscosity.

The values of the average stabilized torque in the interval of 5 to 10 min of the PS, the binary mixtures (PS/PPv and PS/PPr), and the compatibilized mixtures are presented in Figure 5.



Figure 4. Torque curves as a function of PS processing time, binary mixtures, and ternary mixtures.



Figure 5. Average stabilized torque in the range of 5 to 10 min of the PS, the binary mixtures (PS/PPv and PS/PPr), and the compatibilized mixtures.

The average stabilized torque of the PS/PPv mixture was 2.21 N.m, while the PS/PPr system was around 1.6 N.m, lower than the PS (3.7 N.m). The torque reduction was more pronounced in the PS/PPr mixture, indicating that the recycled material works as a diluent and, subsequently, decreases

the viscosity. With the addition and increase of the SEBS content, the stabilized torque increases significantly, suggesting that SEBS is acting as a compatibilizer of the PS/PPv and PS/PPr mixtures, which increases the interfacial adhesion between the matrix and the dispersed phase. A very significant increase is observed for the PS/PP compatibilized blends compatible with 10% SEBS, indicating a greater interfacial improvement of these systems. The increase in the stabilized torque of the mixtures is in line with the trend in impact strength, as shown in Figure 9. Apparently, the compositions PS/PPv/SEBS and PS/PPr/SEBS with 10% compatibilizer have similar torques, because they are within the experimental error range. In this case, even using 20% recycled material, there is a synergistic effect in the mixture and, at the same time, a contribution to cost reduction of the final product. Similar behavior in torque rheometry was verified by Kong et al. [32], indicating that the increase in torque generates gains in impact strength of PP/HIPS/SEPS blends, due to the higher level of interfacial interaction.

3.2. Scanning Electron Microscopy (SEM)

Figure 6 shows the SEM images of the fracture surface of the PS, with a magnification of 1000×. The fracture of the PS presents a smooth and homogeneous aspect, which is associated with the fragile nature of this polymer, corroborating the low impact strength.



Figure 6. SEM image of the polystyrene (PS) fracture surface.

Figure 7 shows the SEM images of the fracture surface of the binary mixtures, with an increase of 5000×. The formation of a biphasic and coarse morphology is observed, with some spherical and elongated particles, of varying sizes, dispersed in the PS matrix. At the same time, there are particles without any type of interfacial adhesion (black circles), indicating a low adhesion between PS and PP, due to the high interfacial tension between the phases, which makes the interface with low resistance [33]. In fact, in Figure 7a,b, empty spaces can be observed, indicating that there was no interaction between PS and PP, as seen in the torque rheometry. The incompatibility between the PS and the PP was corroborated with the results of the impact strengths of these systems, which were lower than those of the compatibilized systems.

Figure 8 shows the SEM images of the fracture surface of the compatibilezd mixtures, with an increase of 5000×. The compatibilized blends, regardless of the SEBS content, showed a more stable morphology, because it significantly reduced the number of voids. Morphology stabilization is an important and necessary requirement for good performance in mechanical and thermomechanical properties [34].

Figure 8a,b shows the fracture surfaces of the PS/PPv/SEBS and PS/PPr/SEBS samples with 5% compatibilizer, respectively. It verifies the presence of PP particles well adhered in the PS matrix, showing that the interface did not break and confirming that the adhesion between the phases was increased, in comparison to systems without compatibilizer. According to Omonov et al. [35], who

used PS as a continuous phase and PP as a dispersed phase in the PS/PP (80%/20%), demonstrated that the presence of compatibilizer can increase adhesion between the phases. In fact, the compatibilizer usually migrates to the interface, increasing the adhesion between the phases, reducing the interfacial tension, and preventing coalescence between the particles of the dispersed phase [36]. However, apparently, part of the compatibilizer migrated to the PS and PP interface, while possibly a small fraction was dispersed in the PS matrix (white arrows). In this case, SEBS is acting as a compatibilizer and impact modifier, as suggested by Mello et al. [37]. The PS/PPv/SEBS mixture (5%) showed relatively larger PPv particles compared to the PS/PPr/SEBS system (5%). Such behavior probably indicates that the lower viscosity of the recycled PP favored a greater interaction with the SEBS, contributing to a greater refinement of the dispersed phase in the PS/PPr/SEBS system (5%). As a consequence, the PS/PPr/SEBS mixture (5%) achieved a more efficient size of recycled PP particles, generating greater energy dissipation and improving the impact strength, in comparison with the PS/PPv/SEBS mixture (5%).



Figure 7. SEM images of the fracture surfaces: (a) PS/PPv; (b) PS/PPr.



Figure 8. SEM images of the fracture surface: (a) PS/PPv/SEBS (5%); (b) PS/PPr/SEBS (5%); (c) PS/PPv/SEBS (10%); (d) PS/PPr/SEBS (10%).

The addition of 10% SEBS significantly reduced the size and improved the dispersion of the PP particles, according to Figure 8c,d. Everything indicates that when a greater amount of SEBS was added it promoted a greater interaction with PPv and PPr, as seen in the torque rheometry. As a consequence, the PS/PPv/SEBS (10%) and PS/PPr/SEBS (10%) mixtures had dispersed particles well adhered to the polystyrene matrix, indicating that there was interaction and the interface did not break. It can be seen in Figure 8c that, in certain regions of the fracture surface, the particles almost touch each other and that, in other regions, there is a great distance between them. On the other hand, in Figure 8d, the PS/PPr/SEBS blend (10%) presented homogeneously distributed PPr particles; i.e., the average distance between the particles is uniform, which favored greater impact strength.

The PS/PPr mixture with 10% SEBS, shown in Figure 8d, presented, in relation to the other systems, a more stable morphology and more refined particles. In this composition, the compatibilizer proved to be effective in improving the properties, as can be seen later in the impact strength and elongation at break. Apparently, for the PS/PPv/SEBS and PS/PPr/SEBS mixtures, the reduction in the size of the dispersed phase within a certain limit is important in the final result, as it increases the interfacial area and improves the transfer of stresses imposed on the system [38,39]. It is reasonable to suggest that the PS/PPr/SEBS mixture (10%) reached the ideal size of the PPr particles, because it promoted the highest toughening and, subsequently, a high performance in impact strength.

3.3. Impact Strength

Figure 9 shows the impact strengths of PS, the binary mixtures (PS/PPv and PS/PPr), and the compatibilized mixtures. The arithmetic mean and the deviation were calculated according to Equations (1) and (2).



Figure 9. Impact strength of PS, binary, and compatibilized blends.

The PS presented impact strength in the order of 20.6 J/m, a value typical of material with fragile behavior [40]. The blends PS/SEBS (5%) and PS/SEBS (10%) were developed, with the aim of

understanding the influence of these levels in the toughening of the PS. The individual addition of 5% and 10% SEBS did not produce a significant increase in the impact strength of the PS, only a slight increase. Therefore, the SEBS compatibilizer is not acting as a polystyrene impact modifier, although the fact that a low concentration is being used must be taken into account.

The direct addition of 30% PPv and PPr to the polystyrene (PS) matrix did not increase the impact strength, only maintaining the fragile behavior. Even though PPv and PPr are copolymers, they are not capable of acting as polystyrene impact modifiers. These results demonstrate the immiscibility and incompatibility of the PS/PPv and PS/PPr mixtures, corroborating the torque rheometry. As there is no interaction between the phases of the PS/PPv and PS/PPr mixtures, the interfacial adhesion is low, as seen in the SEM. In this case, the low interfacial interaction acts as a defect in the PS/PPv and PS/PPr mixtures, contributing to the formation and facilitation of crack propagation, resulting in low impact strength [41,42].

In Figure 9, it is observed that the impact strength of the PS/PPv/SEBS and PS/PPr/SEBS mixtures increased considerably with the increase in the SEBS copolymer content. The improvement of this property is taken as an indicator of the increase in interfacial adhesion between the phases, because SEBS presents part of the chemical structure miscible with PS and PP and, as a consequence, promoted interactions strengthening the system interface [43]. This hypothesis was confirmed by the morphology presented in the SEM. The PS/PPv/SEBS (5%) and PS/PPr/SEBS (5%) blends showed a 45% and 73% increase in impact strength, respectively, compared to neat PS and binary mixtures. When the mixture formulation contains recycled material (PS/PPr/SEBS (5%)), the impact strength tends to maximize; a greater interaction is probably occurring due to the lower viscosity of the PPr. The addition of 10% compatibilizer in the PS/PPv/SEBS mixture promoted an increase of 104% in impact strength, compared to PS. The experiments also showed a considerable increase in impact strength of the PS/PPr/SEBS mixture (10%), reaching a gain of 227% in relation to the PS. Again, the mixture with recycled material demonstrated a tendency to have a greater synergistic effect in this impact property, suggesting that the lower molecular weight of the recycled material favors a greater interaction with SEBS and a plasticizer effect. Apparently, in the PS/PPr/SEBS (5%) and PS/PPr/SEBS (10%) mixtures, the compatibilizer tends to encapsulate the PPr more efficiently, favoring greater molecular entanglement. In general, the SEBS copolymer is effective in making the PS/PP and PS/PPr mixtures compatibilized with the 10% weight content, because it helped to improve the PS toughening, corroborating the trend seen in the torque rheometry.

The increase in impact strength of the PS/PPr/SEBS mixture (10%), with the use of 20% by weight of recycled material, provides an important technological contribution. A major limitation of PS for technological applications is its low impact strength; however, using recycled polypropylene copolymer can extend its application range, provided that SEBS is added as a compatibilizing agent. The impact strength of 67.5 J/m of the PS/PPr/SEBS blend (10%) surpasses some systems in the literature, using a 2.75 J pendulum, such as biopolyethylene [44,45], and homopolymer polypropylene [46,47].

3.4. Tensile Strength

Figure 10 shows the elastic modulus of the PS, the binary mixtures (PS/PPv and PS/PPr), and the compatibilized mixtures. Polystyrene (PS) displayed a higher elastic modulus, indicating high stiffness. The PS/PPv and PS/PPr mixtures lowered the elastic modulus relative to PS, being more pronounced with the recycled material. A reduction of this kind in the elastic modulus can be attributed to the dispersed phase being a copolymer, as well as the fact that it contains rubber in its formulation. Additionally, the low interfacial adhesion, as verified in the SEM, is a factor that contributes to the reduction. The addition of 5% SEBS copolymer promoted a more significant increase in the elastic modulus of the PS/PPr/SEBS system compared to the PS/PPr mixture. However, for the PS/PPv and PS/PPv/SEBS systems, the values remained nearly the same. Again, there are indications that SEBS tends to increase interfacial reinforcement in the mixture with recycled material (PS/PPr/SEBS 5%), as discussed in the section on impact strength. This hypothesis is reinforced when there is a more

significant recovery in the elastic modulus of the PS/PPr system for PS/PPr/SEBS (5%) compared to PS/PPv for PS/PPv/SEBS (5%). The PS/PPv/SEBS (10%) and PS/PPr/SEBS (10%) mixtures displayed a slight increase in the elastic modulus in comparison to those compatibilized with 5% SEBS. Such behavior can be attributed to the increase in SEBS, because it has an elastomeric nature and increases system flexibility.



Figure 10. Elastic modulus under traction for PS, binary mixtures (PS/PPv and PS/PPr), and compatibilized mixtures.

In general, the mixtures did not increase the elastic modulus compared to neat PS. However, the use of PPv or PPr provided results with a good balance between stiffness and impact strength, because there was no severe reduction in the elastic modulus. This is due to the addition of a tough and flexible semicrystalline material (PPv or PPr) to a stiff and amorphous matrix (PS). The mixtures PS/PPv/SEBS (10%) and PS/PPr/SEBS (10%) present important results from a technological point of view, because they can be considered toughened at room temperature (~25 °C). At the same time, PS/PPv/SEBS (10%) and PS/PPr/SEBS (10%) mixtures present satisfactory stiffness for general use applications, because they are at the level of the commercial high impact polystyrene elastic modulus [48].

Figure 11 shows the tensile strengths of the PS, the binary mixtures (PS/PPv and PS/PPr), and the compatibilized mixtures.

Neat PS showed high tensile strength, as it contains the benzene side group in its structure that restricts molecular mobility. The tensile strength of the PS/PPv and PS/PPr mixtures showed the lowest performance compared to neat PS. As it is a property measured in plastic deformation, the low stress causing deformation of the PS/PPv and PS/PPr mixtures was due to poor interfacial adhesion, as verified in the SEM. In addition, PPv and PPr present ethylene/propylene (elastomer) as a dispersed phase, also contributing to reduced tensile strength. In multiphase polymeric mixtures, the factors that directly affect this property, in addition to the individual contributions of each component, are the morphology of the phases present and the adhesion in the interfacial region [49]. In Figure 11, it is observed that the

addition of the compatibilizer SEBS increased the tensile strength for all compositions, indicating that the stress transfer was more effective between the matrix and the dispersed phase. This improvement was due to increased interfacial adhesion and compatibility, as seen in the SEM and torque rheometry. Apparently, there is no significant difference between the mixtures, regardless of whether 5% or 10% SEBS is used, because they are within the experimental error range. Comparatively, mixtures processed with recycled polypropylene have the same level as virgin polypropylene, indicating that it is a good quality raw material.



Figure 11. Tensile strength of PS, binary mixtures, and compatibilized mixtures.

The elongation at break of the PS, the binary mixtures, and the compatibilized mixtures is shown in Figure 12.

The properties of elongation at break is an indicator of ductility under tension; i.e., the degree of deformation. Polystyrene (PS) showed the low deformation typical of a fragile polymer, with low toughness. The PS/PPv and PS/PPr blends presented a deformation similar to that of the PS, because they are within the margin of the experimental error, corroborating the results of impact strength, which were similar. The addition of 5% and 10% of the SEBS copolymer promoted an increasing increase in the elongation at break, suggesting a greater energy dissipation of the compatibilized systems. The increase in this property is a direct indication that there has been an improvement in the phase interface [50], corroborating with the SEM. The PS/PPr/SEBS system (70%/20%/10%) showed a different behavior from the others, indicating that the presence of SEBS in this concentration developed a higher tendency to synergism, with an increase in the order of 123% in the elongation at break, compared to PS.



Figure 12. Elongation at break of PS, binary mixtures, and compatibilized mixtures.

3.5. Heat Deflection Temperature (HDT)

The heat deflection temperature test (HDT) is extremely important for the polymer industry, as it simulates the behavior at temperatures above the environment and, therefore, it is a technique widely used for material selection [51,52]. Figure 13 shows the data obtained for the heat deflection temperature of the PS, the binary mixtures, and the compatibilized mixtures.



Figure 13. Heat deflection temperature of PS, binary, and compatible mixtures.

As shown in Figure 13, the HDT value for polystyrene was in the order of 82.6 °C, a high value because it is a stiff polymer. The mixtures increased the heat deflection temperature by an average of 7.5 °C more than PS. The addition of copolymer polypropylene, regardless of whether it was recycled or not, in an amorphous polymer matrix (PS), contributed to the corresponding increase in thermomechanical strength. The investigated composition range of all mixtures, compatibilized or not, were found to be within the experimental error margin, and thus obtained HDT properties comparable to each other.

The literature [53,54] indicates that the heat deflection temperature increases with the increase of either the degree of crystallinity or the elastic modulus, contributing directly to a greater structural stability at more severe temperatures. Apparently, the HDT of the mixtures was improved by the influence of the crystallinity of virgin and recycled polypropylene, because the neat polystyrene showed the highest elastic modulus and the lowest HDT. The increase in the HDT of the mixtures becomes important from an industrial point of view, because the structural deformation will be kept within acceptable limits after the removal of the molded part at a higher temperature.

The PS/PPr and PS/PPr/SEBS mixtures containing recycled material have the same performance in HDT compared to mixtures with virgin resin, indicating the feasibility of reusing this recycled material. In general, when adding PPv or PPr as impact modifiers to a glassy PS matrix, compared to the styrene-butadiene impact modifier (SBR), it has the advantage of not lowering the heat deflection temperature [55].

3.6. Differential Scanning Calorimetry (DSC)

The results obtained for the melting and crystallization parameters are summarized in Table 2. The DSC curves obtained during the second heating cycle can be seen in Figure 14. In Table 2, it is observed that polystyrene presented a glass transition around 98 °C, compatible with the literature [56]. The glass transition (Tg) of the mixtures, regardless of being compatibilized or not, shifted to higher values, compared to neat PS. Thirtha et al. [57] developed PS/PP mixtures and found an increase in PS Tg. The authors reported that this phenomenon is due to the dispersed particles of PP, which can hinder the molecular relaxation and the mobility of the chain segments necessary for the glass transition process, increasing the Tg. On the other hand, Zander et al. [58] suggested that the increase in PS Tg can be attributed to the molecular confinement of the amorphous domains of PS in the crystalline domains of PP.

Samples *	Tg (°C)	Tm (°C)	Tc (°C)	ΔHm (J/g)	Xc (%)
PS	98.0	-	-	-	-
PS/PPv	106.1	164.1	118.1	12.1	29.2
PS/PPr	106.3	163.9	115.9	17.2	41.5
PS/PPv/SEBS (5%)	106.3	164.0	115.1	14.5	42.0
PS/PPr/SEBS (5%)	107.3	166.6	109.5	10.5	30.4
PS/PPv/SEBS (10%)	107.9	165.2	96.1	5.7	20.6
PS/PPr/SEBS (10%)	107.5	165.5	94.0	10.7	38.7

Table 2. Melting and crystallization parameters for PS, binary and compatibilized mixtures.

* T_g = glass transition temperature; T_m = melting temperature; T_c = crystallization temperature; ΔH_m = melting enthalpy; ΔH_c = crystallization enthaly; X_c = degree of crystallinity, $X_c = \Delta H_m/w*\Delta H_0$, w = weight fraction of PP; ΔH_0 = melting enthalpy of 100% crystalline PP, assumed as 138 J/g [59,60].

In Figure 14a, it can be seen that the mixtures, compatibilized or not, have a single melting peak (Tm), around 165 °C, attributed to the crystalline form, α [60]. The melting temperature of the mixtures did not show significant differences, only a maintenance or a subtle increase in values. On the other hand, the crystallization temperature (Tc) varied more significantly, depending on the composition. The crystallization temperature of the PS/PPv, PS/PPr, and PS/PPv/SEBS mixtures (5%) hardly changed, because the curves are superimposed. However, these compositions have a higher crystallization

temperature, suggesting that injection molded parts can be removed hotter from the molds, without any warping problems, as they solidify at higher temperatures. The PS/PPr/SEBS (5%), PS/PPv/SEBS (10%, and PS/PPr/SEBS (10%) mixtures reduced the intensity of the crystallization peak, suggesting that they need less energy to promote crystallization and, at the same time, shifted to a lower temperature, as shown in Figure 14b. The PS/PPv/SEBS (10%) and PS/PPr/SEBS (10%) mixtures showed a higher reduction in the crystallization temperature, indicating that the high amount of SEBS hinders the crystallization process. As the compatibilizer is amorphous with a complex structure, it may be difficult to crystallize the mixtures.



Figure 14. Differential scanning calorimetry (DSC) curves for PS, binary, and compatibilized mixtures: **(a)** melting temperature and **(b)** crystallization temperature.

The degree of crystallinity varied considerably, with a minimum of 20.6% for the PS/PPv/SEBS mixture (10%), while the maximum value was obtained by the PS/PPv/SEBS system (5%), with 42%. In general, mixtures containing recycled material showed a tendency towards higher crystallinity, with the exception of the PS/PPr/SEBS system (5%), although it still presents a high crystallinity (30.4%). The greater facility to promote a higher degree of crystallinity of mixtures with recycled PP is due to the lower viscosity of this material, as seen in the torque rheometry (Figure 3). Generally, when the viscosity is lower than the crystallizable phase, the alignment of the chains is favored and crystallization occurs in a more orderly manner, increasing the crystallinity fraction [61].

4. Conclusions

Copolymer polypropylene from industrial containers has the potential to be reused as a polystyrene impact modifier as long as it uses a suitable compatibilizer. In this study, the PS/PPr/SEBS mixture (70/20/10 %wt.) showed the greatest synergism of properties, with gains in impact strength, elongation at break, and thermomechanical strength. Additionally, no severe reduction occurred in elastic modulus or tensile strength. The results of the PS/PPr/SEBS mixtures are significant in the area of polymer recycling, with potential for applications in household utensils, CD packaging, toys, and organizing boxes. The results also indicate a good perspective for further studies with this material, with the aim of increasing the content of recycled material in the formulations and, consequently, to minimize harmful environmental effects and add value to a discarded material.

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