

# Toughening of Immiscible rPS/SAN Blends by SEBS Elastomers: Properties and Morphology

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**How to cite this paper:** Bedjaoui, K., Navarro, R., Krache, R., Valentin, J.L., Calderon, R.H. and Torres, A.F. Angel Marcos Fernandez (2022) Toughening of Immiscible rPS/SAN Blends by SEBS Elastomers: Properties and Morphology. *Journal of Materials Science and Chemical Engineering*, 10, 42-62.

<https://doi.org/10.4236/msce.2022.103004>

**Received:** December 23, 2021

**Accepted:** March 25, 2022

**Published:** March 28, 2022

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## Abstract

In this research, an attempt was made to improve compatibility in a polymer blend composed of incompatible constituents, namely, recycled polystyrene (rPS) and polystyrene-co-acrylonitrile (SAN), through the addition of a compatibilizer. The compatibilizing agent, styrene-ethylenebutadiene-styrene block copolymer (SEBS), was added to the polymer blend in ratios of 5 and 10 wt%. For this purpose, blends of rPS and SAN at different ratios, without and with varying concentrations of compatibilizer, were prepared by melt blending using a co-rotating twin-screw extruder. Mechanical properties including tensile and impact strength, rheological properties (RPA), thermal behaviour (DSC) and morphological characteristics (SEM) were evaluated. According to the results obtained by complex viscosity, the blends behave as a pseudoplastic fluid. The results showed that the addition of SEBS increased the Izod impact strength and the elongation at break and decreased the tensile strength and tensile modulus. rPS/SAN blend modified with SEBS had better mechanical properties than the rPS/SAN alloy. SEM photographs revealed that the SEBS was not only distributed in the SAN phase but also distributed in rPS phase in rPS/SAN/SEBS blend. Furthermore, DSC analysis for blends of rPS/SAN gave a good indication of the improvement on miscibility for most compositions. SEM micrographs of tensile fracture surfaces indicated that the formation of the co-continuous phase and the improvement of interface adhesion are the most important reasons for the excellent tensile properties of the rPS/SAN/SEBS blends. Within the range of analysed compositions, the morphologies investigated by SEM are typical of immiscible blends.

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## Keywords

Recycled Polystyrene, SAN, Toughening, Polymer Blends, Rheology

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### 1. Introduction

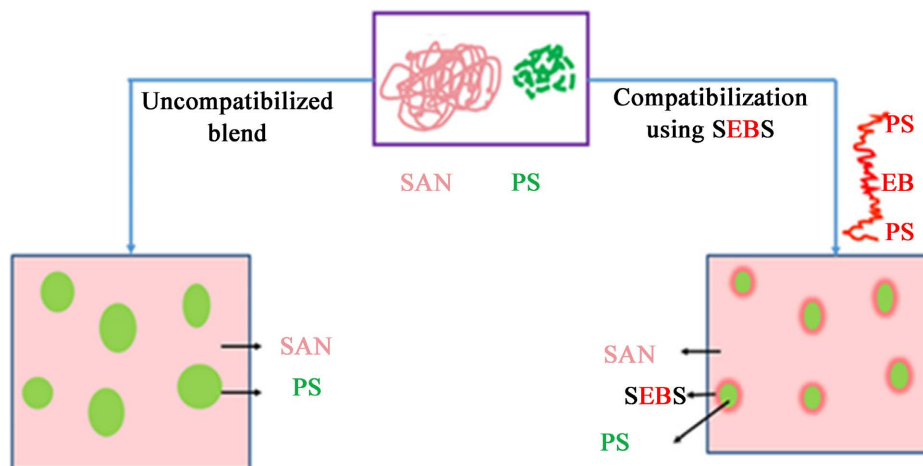
Polystyrene (PS) is one of the most attractive classes of plastics in the world with many valuable properties, Owing to its amorphous structure, is used in various technological applications such as hardness, recyclability, ease of fabrication and lower price compared to other polymers. These characteristics make it very useful for packagings, automotive applications, food containers. On the other hand, styrene-co-acrylonitrile SAN is a widely used engineering thermoplastic due to its desirable properties, which include high impact properties, chemical resistance and easy processing characteristics. The benefits of these polymer properties have led to the wide range of usage especially, the fittings of mobile industry and home appliance, instrument panel, various switches and lamina of the fan, etc. Furthermore, SAN is the matrix phase of the multiphase ABS. However, PS wastes are not degradable, which damages the ecosystem, so the large amount of PS plastic wastes produced nowadays makes it imperative to search for recycling them [1] [2] [3] [4]. Currently, the recycling ways mainly include the reuse of the original plastic waste, mechanical recycling [5] [6] and chemical recycling [7]. Mechanical recycling is a good option from both economic and environmental points of view. However, the recycled polystyrene (rPS) has unsatisfactory mechanical properties of low impact strength because of the degradation during reprocessing.

Blend of rPS/SAN are considered as one of the most important commercial engineering thermoplastic blends since they combine the desirable advantages properties of rPS and SAN. These advantages include the good thermal stability and impact toughness of SAN and low cost [8], hardness and clarity of rPS [9] [10]. However, blends of rPS and SAN are immiscible throughout the whole range of composition [11]. And exhibits poor interfacial adhesion which causes heterogeneous morphology, weak dispersion, phase separation and poor mechanical properties [12] [13] [14]. Therefore, Compatibilization of a polymer blend is often carried out by the introduction of block or graft copolymers. These copolymers concentrate at the interface between the polymeric phase domains, reducing the interfacial tension between the phases and suppressing coalescence [15] [16] [17].

A common approach to improve the toughness of incompatible blends is chemical or physical compatibilization [18] [19] [20]. Therefore, a compatibilizer is needed to improve the interfacial tension and dispersion of the immiscible polymer blends. It is well-known that copolymers, especially block copolymers such as styrene-ethylene-butylene-styrene (SEBS) are effective for this purpose [21] [22]. Yang *et al.* [23] studied the miscibility of polystyrene/poly (styrene-co-

acrylonitrile) (PS/SAN) blend, one kind of A/A-B type blend. The calculation results from these studies suggested that PS/SAN blend shows the Upper Critical Solution Temperature (UCST) behavior within the AN's volume fraction range of 4% - 20%. PS/SAN blend was then studied with Small Angle Light Scattering method (SALS) to test the theoretical prediction. It showed that PS/SAN (AN: 20 vol.%) blend (49/51wt%) has UCST behavior with the UCST temperature of 171.8°C. S. Joseph *et al.* [24] investigated the morphology and mechanical properties of polystyrene (PS)/polybutadiene (PB) blends in the presence and absence of random and tri-block copolymers of PS and PB. Recently introduced rigid-rigid polymer toughening idea has provided a new route for improving the mechanical properties. Thus, it has attracted considerable attention because of its industrial importance among other types of polymer blends. [25] [26] and extend their application range so far [27]. While, a large number of toughened polymers such as high impact polystyrene (HIPS) and acrylonitrile-butadiene-styrene copolymer (ABS) have been developed and widely employed as engineering materials [28]. Cavanaugh *et al.* [29] observed an enhancement of 800% in the impact resistance of polystyrene (PS)/polybutadiene blends (23 vol% rubber) compatibilized with a PS-polybutadiene block copolymer in comparison with PS. This enhancement was attributed to the long asymmetric chain of the diblock compatibilizer, which was capable of entangling in both homopolymer phases. However, it is still necessary to understand the relationship between rheology, morphology and mechanical properties of rPS/SAN blends to propose mechanisms to compatibilization (Figure 1). Of the polymeric system and improve the mechanical performance of the recycled material. In literature, there is no information that reports about the compatibilization of rPS/SAN blends and evaluates the effect of SEBS as compatibilizer on the morphology on mechanical and rheological properties of recycled PS with SAN. Hachiya *et al.* [30] indicated that PS is partly miscible with SAN containing less than 5 wt% acrylonitrile.

In summary, an improved toughness of immiscible rPS/SAN blends due to the addition of suitable triblockter polymers without decreasing much of its tensile



**Figure 1.** Schematic representation of morphology of SEBS compatibilized blend.

property has been confirmed; however, a more detailed investigation of the rheological phenomena related to the elastic as well as inelastic deformation behaviour of such rPS/SAN/SEBS blends is desirable. The aim of this work, therefore, was to establish a systematic correlation between the blend rheological, mechanical and morphological behaviour of uncompatibilised as well as compatibilised rPS/SAN blends.

## 2. Experimental Part

### 2.1. Materials

The recycled Polystyrene (rPS) used in this study was the waste collected from moulding that was grinded with a mill), were obtained from industrial containers gathered by the company IRIS 7-(Algeria), with a melt flow index (MFI) of 25 g/10min at 200°C, 5 kg).

The materials used in this study were SAN (Luran 368R), with 22 wt% of (AN), ((MFI = 11 g/10min) at 190°C) and with a density of 1.08 g/cm<sup>3</sup>. Supplied as pellets and manufactured by Basf, Germany. The properties of rPS and SAN respectively are shown in **Table 1**.

The SEBS triblock copolymer used was Kraton G1651 obtained from Shell chemical Company. Contained 72 wt% (EB) random copolymer and 28 wt% of styrene.

### 2.2. Blend Preparation

Before use, all the pure polymers and their blends were dried: the rPS and the SAN, at 100°C under vacuum oven for 24 h, and the SEBS at 70°C respectively. Preliminary experiments were carried out in a PlasticorderBrabender (LP2100, Duisburg), Germany allowing to record the torque vs time at a temperature of 200°C during 10 min cycle with 50 rpm screw speed. The obtained samples were cut into small pieces and then compression moulded into 2-mm-thick sheets and thin films at the same temperatura (200°C) for 8 min using a hydraulic press at a pressure of 75 Pa. Subsequently, the composition of blend without and with SEBS are summarized below in **Table 2**.

## 3. Blend Characterizations

### 3.1. Rheological Properties

Rheological measurements were performed using RPA 2000 Rheometer (Alpha

**Table 1.** Properties of the recycled PS and SAN grades used for the uncompatibilized and compatibilized rPS/SAN blends.

	$T_g$ (°C)	$T_m$ (°C)	Mn (g/mol)	Mw (g/mol)	Mw/Mn
rPS <sup>a</sup>	93	-	87,000	300,000	3.4
SAN	111	-	43,000	121,000	2.6

<sup>a</sup>Tradename: GPPS code 1540 (Total petrochemical, Belgium).

**Table 2.** The experimental compositions of rPS, SAN and their blends.

rPS/SAN	Without SEBS	SEBS (5%)	SEBS (10%)
100/0	rPS	-	-
30/70	rPS30	rPS30C5	rPS30C10
50/50	rPS50	rPS50C5	rPS50C10
70/30	rPS70	rPS70C5	rPS70C10
0/100	SAN	-	-

Technologies) at 190°C. Frequency sweeps were carried over a frequency range of from  $10^{-2}$  to  $10^2$  Hz with strain amplitude ( $\gamma = 0.02$ ).

### 3.2. Mechanical Properties

The mechanical properties, including (tensile strength, elongation at break and Young's modulus of rPS/SAN) were measured using a Instron 3366 (USA) tensile tester at room temperature ( $23^\circ\text{C} \pm 1^\circ\text{C}$ ) with a crosshead speed of 10 mm/min. The shaped specimens and the measurement were carried out according to ISO 527-2. Five specimens were tested for each reported value.

Notched Izod impact tests were performed on injected samples (thickness 1.9 mm) using a resilipector apparatus (Ceast 6548) at  $25^\circ\text{C}$ . The notches (depth 1.54 mm) were machined after injection molding. A minimum of seven impact specimens were tested for each reported value, and the average values were calculated then reported [4].

### 3.3. Differential Scanning Calorimetry (DSC)

The glass transition temperature ( $T_g$ ) was determined by DSC (METTLER-TOLEDO DSC 822° under nitrogen atmosphere. The weight of sample capsule was (9 - 15 mg). The samples were heated from  $25^\circ\text{C}$  to  $150^\circ\text{C}$  at a rate of  $20^\circ\text{C}/\text{min}$  to eliminate previous thermal history and were held at this temperature for 2 min to ensure complete melting before starting the cooling.

Cooling was carried out from  $150^\circ\text{C}$  to  $-90^\circ\text{C}$  at  $10^\circ\text{C}/\text{min}$  followed by heating from  $-90^\circ\text{C}$  to  $150^\circ\text{C}$  at  $10^\circ\text{C}/\text{min}$ .  $T_g$  was taken as the midpoint of the transition. The occurrence of a single  $T_g$  was taken as confirmation of miscibility.

### 3.4. Scanning Electron Microscopy (SEM)

Cryogenically fractured surfaces of PS/SAN blends specimens were observed, after gold-coating, by scanning electron microscopy using a Hitachi SU800 (Japan) electron microscope operated at 15 kV.

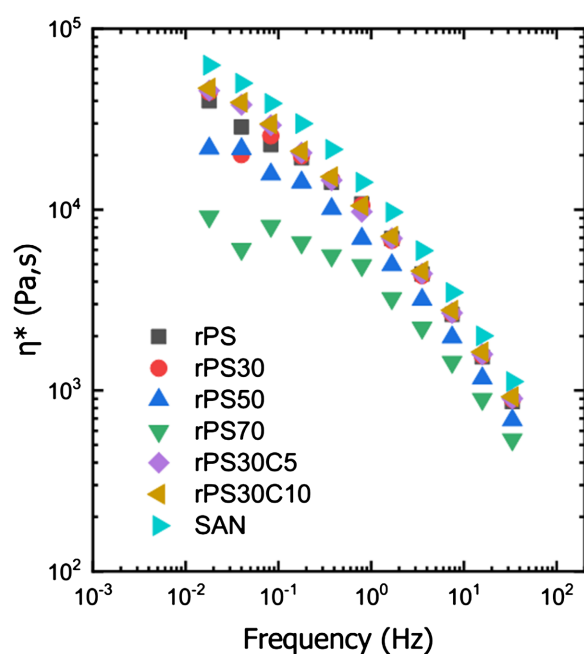
## 4. Results and Discussion

### 4.1. Rheology Behavior of the Polymer Blends

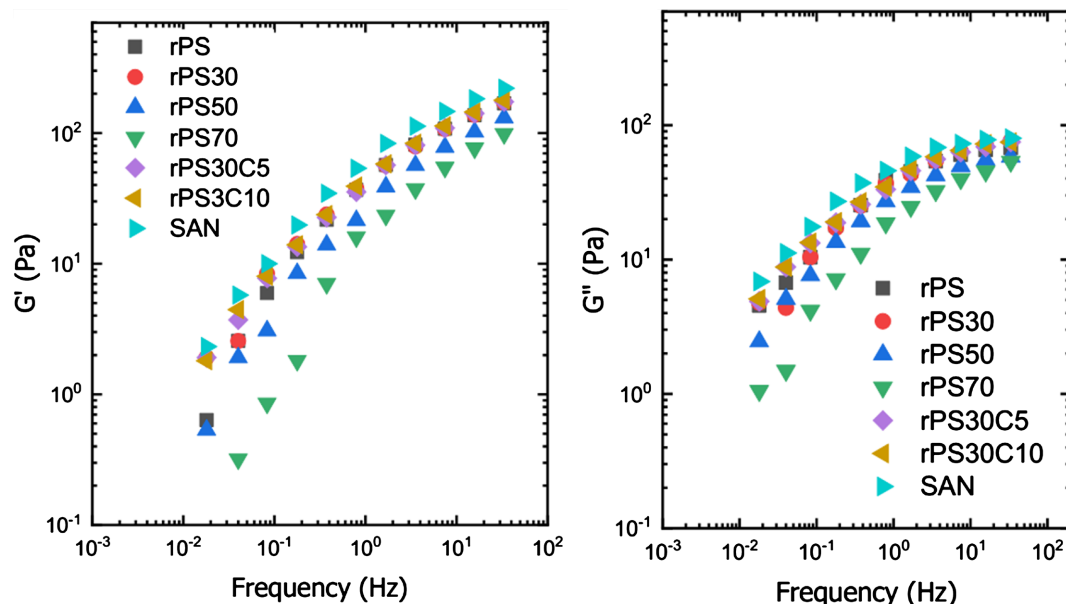
The results of the rheological measurements carried out on the blends are shown in **Figures 2-5**, where their viscosity curves are plotted as a function of frequen-

cy (together with those of the pure components for the pure rPS, SAN and their blends at 190°C in the 100 - 0.01 Hz as a function frequency range, which revealed their complex viscosity ( $\eta^*$ ), storage moduli ( $G'$ ) and loss moduli ( $G''$ ). However, Rheological properties provides information about the compatibilization effect, as well as the correlations between their rheological–morphological–mechanical properties [31].

**Figure 2** shows the changes in complex viscosity ( $\eta^*$ ) as a function of angular frequency ( $\omega$ ) of rPS, SAN, and their rPS/SAN blends without and with 5 wt% and 10 wt% of SEBS. It can be observed that the complex viscosity of the SAN was higher than that of the recycled PS. In addition, **Figure 2** also shows the effect of SEBS copolymers with different ratios 5 wt% and 10 wt%, on the complex viscosity of rPS/SAN blends. The complex viscosity of all formulations shows a downward trend with increasing the frequency, reflecting the shear thinning phenomenon and exhibiting the typical motion characteristics of pseudoplastic fluid [32]. This is because the molecular chain entanglement decreases with increasing the shear force, leading to a better fluidity of blends. However, compared with pure rPS/SAN blends, the complex viscosity of the blends increases a little bit after adding SEBS copolymers which is probably due to the physical compatibilization of SEBS and may be due to a low fluidity of SEBS when compared to rPS and SAN, or to the interactions performed during blending, between styrenic and EB block molecular segments of SEBS with rPS and SAN, respectively. Suggesting the changes have occurred in the morphology and interface of compatibilized blends [31], which may directly impact the mechanical properties. This assumption correlated with the results obtained with mechanical and SEM images properties in the following section discussed.



**Figure 2.** Effect of Complex viscosity ( $\eta^*$ ) of rPS, SAN and their blends.



**Figure 3.** The Frequency-sweep plot of the storage modulus ( $G'$ ) and loss modulus ( $G''$ ) for neat rPS, SAN and uncompatibilized/compatibilized rPS/SAN/SEBS blends.

The rheological measurements were carried out, The relation curves of storage modulus ( $G'$ ) and loss modulus ( $G''$ ) of rPS/SAN blends as a function of the frequency are shown in **Figure 3(a)** and **Figure 3(b)**, respectively. The  $G'$  and  $G''$  of all samples are observed to increase with increasing the frequency. In the low frequency region, the movement of molecular chain is synchronized with the change of external force, and the flexibility and elasticity of molecular chain are high, which result in a lower  $G'$  of the blends. Meanwhile, the smaller friction between the molecules causes the lower  $G''$ . In the high frequency region, the movement of molecular chain could not keep up with the change of external force, and the friction consumption between the molecular chains is also increased, which leads to the increase of  $G'$  and  $G''$  [32].

It can also be seen that the storage modulus ( $G'$ ) and loss modulus ( $G''$ ) of the rPS/SAN blend compatibilized with SEBS copolymers are higher than those of the uncompatibilized rPS/SAN blends, indicating an increase of the interaction between rPS and SAN on addition of SEBS compatibilizer. The rheological properties in low frequency region can be considered to reflect the relaxation and motion of the whole polymer chains. Also the  $G'$  and  $G''$  of the blends reach the maximum value at 10 wt% of SEBS, which is due to the chain entanglement between SEBS and the blends. On the other hand this increase in  $G'$  and  $G''$  is associated with a relaxation time of dispersed phase [33] [34].

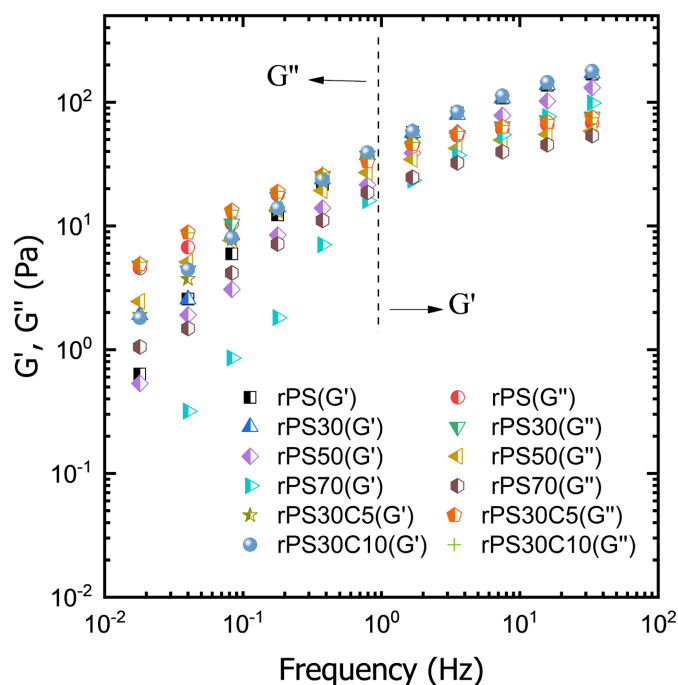
As well mentioned in literature [31] the storage modulus of an immiscible blend is characterized by the presence of a relaxation shoulder, which arises due to deformation and relaxation of dispersed phase, this trend is verified for all blends analyzed. Compared to binary blends (rPS/SAN),  $G'$  of SEBS compatibilized blends is higher at low frequencies, indicating higher elasticity, possibly this increase is related with SEBS addition, these  $G'$  data strengthen the assump-

tions that SEBS is acting as a compatibilizing agent, favoring interactions between chemical groups [34]. Which presents high viscoelasticity, as well as with dispersed phase reduction particle size,

The relation curves of storage modulus ( $G'$ ) and loss modulus ( $G''$ ) rPS/SAN blends as function of the frequency at 190°C are shown in **Figure 4**. It is observed that, at lower frequencies below 1 Hz, the viscous response is higher than the elastic response for all samples analysed, until reaching a frequency, where the modules intersect  $G' = G''$  (where the dissipative feature is equal to the elastic feature at the same frequency). For values higher than 1 Hz, the elastic response ( $G'$ ) exceeds the viscous response ( $G''$ ).

According to Bousmina and Muller [35], they reported similar behavior for other copolymers containing rubber. In these studies, the  $G' = G''$  cross-points were attributed to network type structures formed through the association of elastomeric particles. While this inversion of behavior is associated with hindrance in molecular mobility, since the molecular segments can not move and respond as rapidly as the applied frequency. **Table 3** shows values for the point  $G' = G''$ , for all investigated compositions.

The rubbery phase causes differences in the  $G' = G''$  cross-points of the copolymers SAN (where the dissipative feature is equal to the elastic feature at the same frequency), when compared with those of the homopolymers rPS at 190°C. In the homopolymers and in SAN, the  $G' = G''$  crosses occur at high frequencies and only at single points. SAN at 190°C exhibits a band of  $G' = G''$  crosses (**Figure 4**), rather than a single angular frequency point as was observed for PMMAh, PS, and SAN [36]-[42].



**Figure 4.** The relation between storage modulus ( $G'$ ) and loss modulus ( $G''$ ) of rPS/SAN blends as function frequency for all compositions analysed.



**Table 3.** Crossing-point,  $G' = G''$ , for rPS and their blends.

Composition	Frequency (Hz)	$G' = G''$ (MPa)
rPS	0.77	38.11
rPS30	0.80	37.03
rPS50	1.67	37.10
rPS70	3.52	35.56
rPS30C5	0.79	34.07
rPS30C10	0.79	36.48

## 4.2. Mechanical Properties

### 4.2.1. Impact Strength of Recycled rPS/SAN Blends

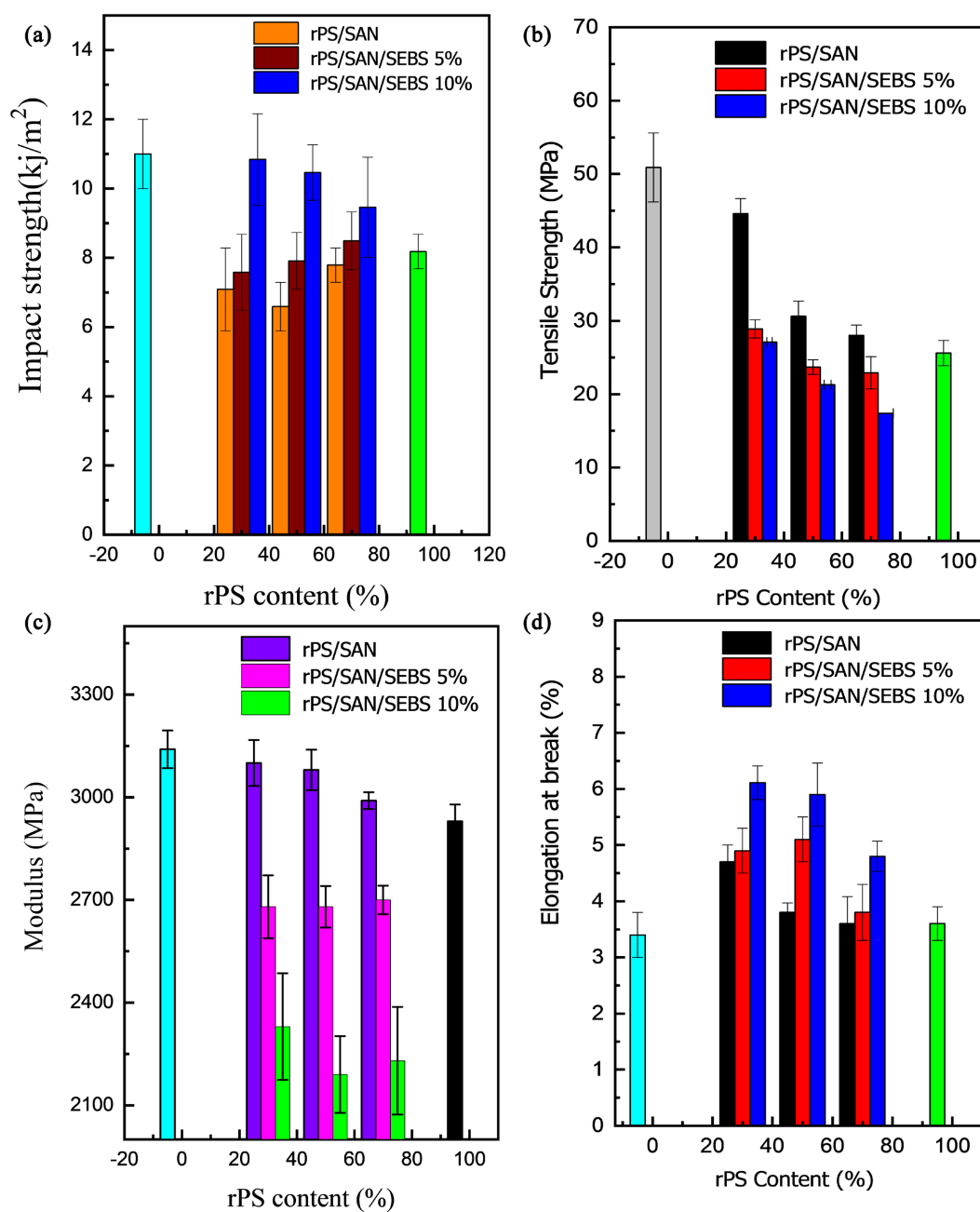
The notched Izod impact strength is one of the most important properties for rigid polymers such as the rPS/SAN alloy because of their notch sensitivity (**Figure 5(a)**), shows that SEBS is a good compatibilizer and tremendously improves the impact strength of the rPS/SAN with a SAN content 30 wt% - 70 wt%.

The impact strength of the rPS/SAN alloy can be greatly enhanced by the addition of 5 wt% and 10 wt% SEBS. The impact strengths of simple rPS/SAN blends without SEBS are very low, indicating a negative synergistic effect between the rPS and the SAN because of their poor interfacial interaction in the rPS/SAN blends.

As well known that the phase morphology and the interfacial adhesion between component polymers influence the mechanical properties of polymer blends. Two-phase morphology with lack of adhesion between the component polymers leads to premature failure and, thus, to poor mechanical strength, and the improved interfacial adhesion leads to higher mechanical strength. The izod impact strength of rPS, SAN and rPS/SAN compatibilized blends is shown in **Figure 5(a)**. It can be seen that the addition of SEBS to rPS/SAN blend results in better impact strength. For example, the impact strength increases from 7.1 kJ/m<sup>2</sup> of rPS<sub>30</sub> to 10.8 kJ/m<sup>2</sup> when 10 wt% SEBS is added. However, the impact strength becomes to decrease with the further increasing of SAN content up to 70 wt%, this is probably due to the poor compatibility between SAN and rPS and may be also attributed to its brittle behavior.

According to **Table 4**, it can be observed that the impact strength of the SAN is higher than that of recycled PS and the impact strength of the uncompatibilised rPS/SAN blends shows lower value than the pure polymers. In fact, the low impact strength of rPS/SAN blends may be related to their poor adhesion and weak interfacial interaction between the two phases caused by incompatibility between recycled PS and SAN, as further shown in SEM images.

In other hand, the incorporation of SEBS can improve the compatibility of the rPS/SAN blends. Moreover, the impact strength, of the binary rPS/SAN blends increases with increasing SEBS content (which acts as an impact modifier that improves the toughening and ductility of the brittle rPS/SAN, thus increasing



**Figure 5.** (a) The Notched Izod impact strength of rPS/SAN blends without and with SEBS compatibilizer as a function of rPS content; (b) The Tensile strength of rPS/SAN blends without and with SEBS compatibilizer as a function of rPS content; (c) The Tensile Modulus of rPS/SAN blends without and with SEBS compatibilizer as a function of rPS content; (d) The Elongation at break of rPS/SAN blends without and with SEBS compatibilizer as a function of rPS content.

the energy absorption capacity of the blends. When highly dispersed, the SEBS rubbery phase acts as an effective stress concentrator, thus enhancing both crazing and shear yield in the blend. Because both processes can dissipate a large amount of energy, there is a significant increase in the toughness of the blends. This increase of impact strength is possible due to the fact that SEBS compatibilizer contains (Eb) block segments chemically identical to rPS and with SAN,

**Table 4.** The mechanical properties of recycled PS, rPS/SAN, and rPS/SAN/SEBS blend.

rPS/SAN	Tensile Strength (MPa)	Tensile Modulus (MPa)	Elongation at break (%)	Impact Strength (kJ/m <sup>2</sup> )
rPS	25.6 ± 1.7	2930 ± 48	2930 ± 48	8.2 ± 0.5
rPS30	44.6 ± 2	3100 ± 67	3100 ± 67	7.1 ± 1.2
rPS50	30.6 ± 2	3080 ± 59	3080 ± 59	6.6 ± 0.7
rPS70	28.7 ± 1.4	2990 ± 24	2990 ± 24	7.8 ± 0.5
rPS30C5	28.9 ± 1.2	2680 ± 61	2680 ± 61	7.6 ± 0.1
rPS50C5	23.7 ± 1	2680 ± 90	2680 ± 90	7.9 ± 0.8
rPS70C5	23 ± 2.2	2700 ± 41	2700 ± 41	8.5 ± 0.8
rPS30C10	27 ± 1	2330 ± 155	2330 ± 155	10.8 ± 0.3
rPS50C10	21.3 ± 1.2	2190 ± 112	2190 ± 112	10.4 ± 0.8
rPS70C10	17.4 ± 2.6	2230 ± 157	2230 ± 157	9.5 ± 0.4
SAN	51 ± 4.7	3140 ± 55	3140 ± 55	11 ± 1

which have relatively high affinity with the SAN phase.

It concludes that the SEBS acts as an efficient compatibilizer, this finding reinforces the hypothesis of properly interaction between the elastomeric block (EB) with SAN while terminal groups of styrene with recycled PS [43] [44] [45] [46].

#### 4.2.2. Tensile Strength of rPS/SAN Blends

Generally, it has been long established that immiscible polymer blends have inferior mechanical properties because of the existence of weak interfacial adhesion and poor dispersion of the components. In this study, the mechanical properties before and after the compatibilization effect of rPS/SAN blends by the SEBS block copolymer were investigated.

In terms of tensile (strength, Modulus and Elongation at break) as a function of recycled PS, SAN and the SEBS blends are summarized in **Table 4**. It can be seen that the mechanical properties of SAN are higher than those of rPS.

The tensile strength of the blends with a varied ratio of SEBS is different from the notch impact strength, which can be observed in **Figure 5(b)**. The tensile strength, mixed with 5 wt% and 10 wt% SEBS decreases slightly as compared with pure recycled PS, SAN and rPS/SAN blends. This may be attributed to the fact that SEBS is an elastomer with a lower tensile modulus and affects the rigidity of the blends. With the addition of the same mass fraction of SEBS copolymer.

As expected the reduction in tensile strength of the blends during the mechanical test implies a higher energy dissipation, also this associated with two hypotheses; the SEBS elastomeric character that heads to more flexible blends requiring lower tensile load for deforming; or due to interfacial saturation due to compatibilizer excess, this approach corroborates impact results, whereas the compatibilizer content increases the greater the impact strength, *i.e.*, with higher dissipation energy level for the blends, while reducing the tensile strength compared to neat SAN. It is common knowledge that the toughness will be in-

creased, whereas the modulus will be decreased when elastomer is added [47].

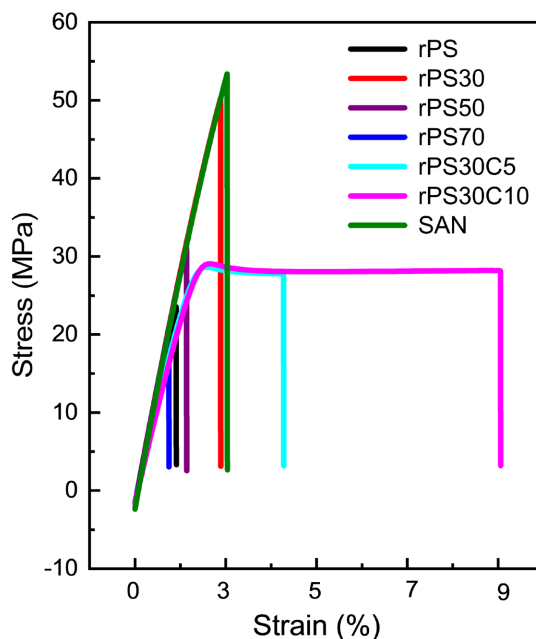
**Figure 5(c)** shows the evolution of elastic modulus of recycled PS, SAN and rPS/SAN blend as function of both rPS and SEBS content. It can be seen that SAN had the highest elastic modulus, with a typical brittle behavior of rPS/SAN blends. Compatibilized blends had reduced elastic modulus in relation to pure SAN and rPS. The decrease in the modulus with increasing elastomer content is expected and well reported for the rubber toughening of rigid polymers [48]. As revealed in literature by la Mantia *et al.* [49]. The SEBS is able of reducing the brittleness of at least one of the two phases and locates selectively at the interface, it was also verified that SEBS decreased the elastic modulus of blends, this being attributed to the SEBS's elastomeric nature. However, these losses in the elastic modulus were accompanied by gains in impact strength, as shown in **Figure 5**.

The elongation at break of the binary rPS/SAN blends in comparison with neat rPS, SAN and ternary rPS/SAN/SEBS blends is shown in **Figure 5(d)**. Addition of SEBS, the softness and flexibility of this polymer will increase the elongation of the blend. All values are higher than that of the pure SAN.

As expected, it can be seen in **Table 3** that the most effective compatibilizer was a styrene-ethylene/butadiene-styrene triblock copolymer (SEBS). This substance caused a significant increase in the ductility of rPS/SAN. Even at as low as 5 wt% compatibilizer, the elongation-values increased over all samples compatibilized. This is considerably higher than has previously been reported for PS/SAN blends compatibilized by SEBS [46]. At least two explanations exist for this effect of the SEBS. Firstly, the SEBS is a block copolymer with sections that are similar and, thus, compatible, to the two types of polymers (polystyrene and styrene-co-acrylonitrile) in the blend. It could act as a surface-active material, reducing the surface tension between the two phases during processing. This reduction in interfacial tension should result in a reduction of the domain size can be expected to improve the properties of the mixture [46].

An alternative explanation offered by La Mantia is that the SEBS is not a true compatibilizer, instead it reduces the brittleness of at least one of the phases instead of accumulating at the interfaces [49].

**Figure 6** shows the tensile stress-strain curves of the pure recycled PS, SAN, and the rPS/SAN blends containing different amounts (5 wt% and 10 wt%) of SEBS. It can be seen that rPS and SAN copolymer present hard, brittle and fragile character with an ultimate elongation less than 4%. However, the blend of Recycled PS and SAN without addition of SEBS does not display any improvement in toughness. It is still brittle and fractured in a brittle mode (**Figure 6**). There is no yield point on its stress-strain curves and it broke at a strain of about 3.5%. On the other hand, the rPS/SAN blend combines the poorest properties of the components. However, after the addition of only 5 wt% of SEBS to the blend, a completely different tensile behavior occurs (**Figure 6**). (Curve of rPS30C5), the blends display a ductile behavior as indicated by the presence of a yielding point. Its stress-strain curve exhibits the characteristics of a typical toughened plastic.



**Figure 6.** Stress *versus* strain plots of rPS, rPS/SAN blend and compatibilized ones as function of rPS content.

Such as acrylonitrile-butadiene-styrene terpolymer and high impact PS. All rPS/SAN blends compatibilized by SEBS also exhibit a similar behavior. The tensile bars of these blends show whitening but no obvious necking in the tensile process. Indicating its ductility and high stiffness while the fragile character was maintained, corroborating the results of impact strength, which were similar.

In contrast, the incorporation of the third components SEBS with 5 wt% and 10 wt% showed higher strain compared to SAN, rPS and rPS/SAN blends, and changed the brittle fracture to a ductile fracture. This increase in strain and absorption of more energy indicates an increase in flexibility of the compatibilized samples in comparison to samples rPS, SAN and rPS/SAN. It is noteworthy that the increase in flexibility in samples is compatibilized to be compared to samples without SEBS, which is consistent with the results of impact strength already presented. It may be noted that the compatibility of blends was improved upon SEBS addition increase for all samples analysed, which resulted in a gradual increase of elongation at break. Similar results were reported in the literature. Diaz *et al.* [48] reported that the increase in elongation at break due to the adhesion between matrix and dispersed phase. It is important to note that an increase in toughness is normally associated with a decrease in stiffness and strength [46], in our case the addition of the SEBS only caused a moderate decrease in these important mechanical properties.

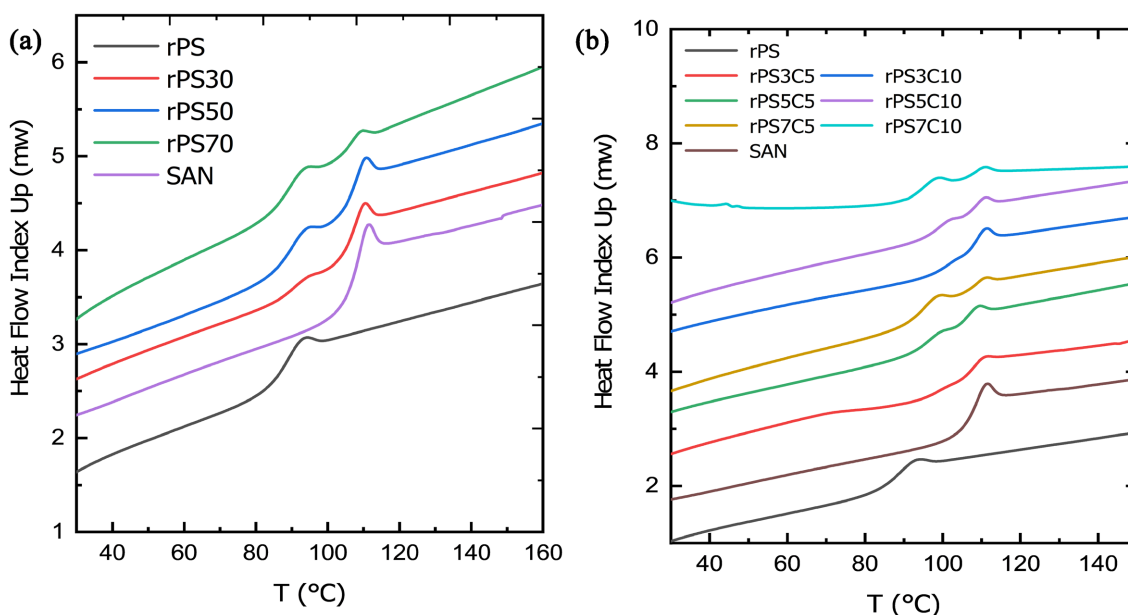
### 4.3. DSC Analysis

The glass transition behavior of rPS, SAN, and their blends, without and with SEBS as compatibilizer, at different concentrations was studied by DSC (**Figure 7(a)** & **Figure 7(b)**). The results obtained on samples with 30 wt%, 50 wt%, and

70 wt% rPS are summarized in **Table 5**. It can be seen that the  $T_g$  values of rPS and SAN determined were 89.04°C and 110°C, respectively. The range of testing temperature was from 0 to 150°C, the  $T_g$  of the EB part in SEBS was not included in here.

And the  $T_g$  of the (St) part in SEBS was overlapped by the  $T_g$  of SAN. However, as SEBS was added into rPS and SAN, their  $T_g$  changed to 98.83°C and 107.53°C, respectively. Two  $T_g$ 's occurred in the rPS/SAN alloy, namely, a  $T_{g2}$  of 106.33°C for SAN phase and the other  $T_{g1}$  of 89.93°C for rPS30 phase. There were also two  $T_g$ 's in rPS50 and rPS70 alloy. Moreover, all the blend compositions show two distinct  $T_g$ 's corresponding to neat rPS and SAN phases, suggesting the immiscible nature of the blend. Generally, the decrease in the difference of the glass transition temperatures of the two phases implies an improved compatibility in the polymer blends.

The  $T_g$  difference ( $T_{g2} - T_{g1}$ ) in the rPS/SAN/SEBS blends is lower than that of the rPS/SAN blends, which confirms that the SEBS can enhance the compatibility



**Figure 7.** Dsc thermograms of rPS, SAN and their blends without and with SEBS.

**Table 5.** The Glass Transition Temperatures ( $T_g$ ) of rPS, SAN, and their blends.

Samples	Ratio	$T_{g1}$ (°C)	$T_{g2}$ (°C)	$T_{g2} - T_{g1}$ (°C)
rPS	100	89.04		
SAN	100	-		
rPS/SAN	30/70	89.93	106.33	16.4
	50/50	89.96	107.23	17.27
	70/30	89.56	104.75	15.19
	30/70/5	98.83	107.53	8.7
rPS/SAN/SEBS	30/70/10	101.12	107.59	6.47

of the rPS/SAN blends. So, it could imply that SEBS was not only distributed in SAN phase but also distributed in rPS phase. This implication could be proved by morphology observation as follow.

#### 4.4. SEM Study

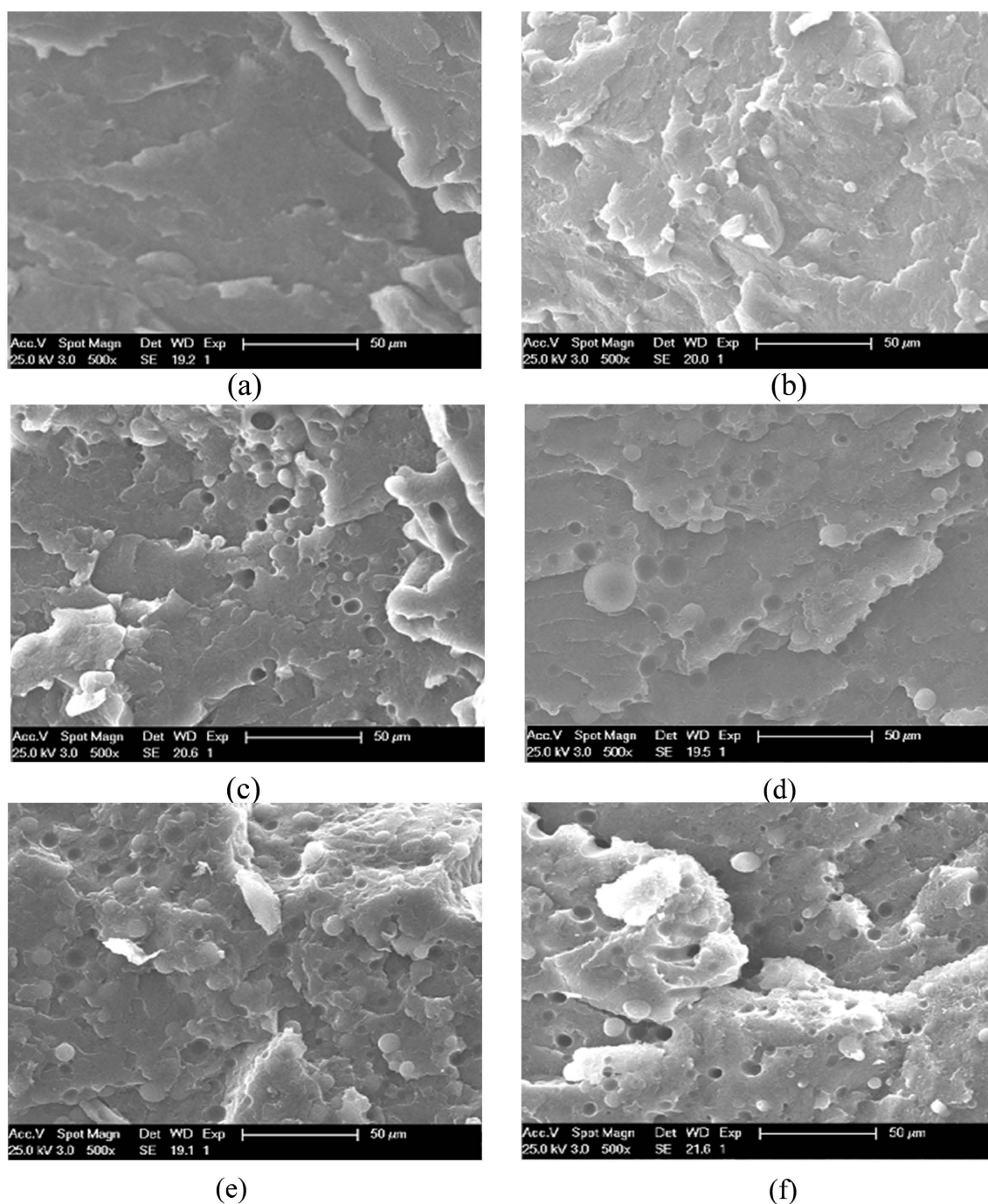
The phase morphology of immiscible polymer blends depends on several factors including composition, processing conditions, interfacial tension, and rheological properties of the individual constituents. The micrographs of the cryogenically fractured surface of rPS/SAN and rPS/SAN/SEBS compounds are displayed in **Figures 8(a)-(f)**, with major content of SAN (rPS30) and major content on rPS (rPS70), respectively. By analyzing morphology of physical blends, it is possible to note that in both cases there are fragile-ductile fracture edges and wide distribution of domain sizes. The inspection of these micrographs indicates two phases with different domain sizes and shapes, which reveal immiscible character of rPS/SAN, with voids presence and dispersed particles.

It is well known that the polymer present in the lower concentration usually forms a discontinuous phase whereas the polymer present in the higher concentration forms a continuous phase and such morphology of blends is usually named a particle-in-matrix type. In addition, low interaction is noted, which suggests poor adhesion between recycled PS and SAN, due to the weak interfacial interaction among the phases in the blends. In general, high interfacial tension of the blends with poor compatibility will cause poor transformation of load from matrix to the dispersed phase, leading to a smooth morphology.

In compatibilized blends with 5% and 10% of SEBS can be clearly observed a size reduction of dispersed particles in recycled PS matrix compared to the binary blend, this result agrees with literature [49] [50]. This effect is related to the compatibilizer migration to the blend interfaces, promoting a reduction of interfacial energy and avoiding coalescence [44]. The compatibilizer diffusion to the interface is reinforced as observed in **Figure 8**, which indicates the prominent compatibilization of the SEBS. The possible reasons are that SEBS facilitates the dispersion of the recycled PS dispersed phase and results in strong interfacial adhesion of the rPS/SAN blends.

**Table 6.** Average size of holes in SEM morphology of the blend analysed with ImageJ software.

	$R_n$ ( $\mu\text{m}$ )	$R_w$ ( $\mu\text{m}$ )
rPS <sub>30</sub>	2.52	4.18
rPS <sub>30</sub> C5	3.4	5.86
rPS <sub>30</sub> C10	1.37	3.35
rPS <sub>70</sub>	2.89	3.14
rPS <sub>70</sub> C5	5.60	5.71
rPS <sub>70</sub> C10	7.43	6.53



**Figure 8.** SEM micrographs of (a) rPS30; (b) rPS30C5; (c) rPS30C10; (d) rPS70; (e) rPS70C5; (f) rPS70C10 blends.

In conclusion, compatibilizers play a key role to improve the interfacial adhesion between the components and to reduce the interfacial tension between the components. They exhibit interfacial activities in heterogeneous polymer blends. The interfacial activities of compatibilizers help to stabilize the morphology by enhancing interfacial adhesion. Compatibilizers resist the coalescence of dispersed phases, thereby reducing the interfacial tension and the size of the dispersed domains which results in an increase of adhesion at the interface and im-



proved properties of the final product. Commonly used compatibilizers are block, graft, or random copolymers consisting of dissimilar blocks [51].

## 5. Conclusion

In this research, the rheological, mechanical and morphological properties of the recycled PS rPS/PSAN and rPS/SAN/SEBS blends were studied. The addition of SEBS copolymers increased the viscosity of all compositions at low frequencies suggesting that the reactions between the copolymers and blend were achieved. The complex viscosity increased with the increase in the content of copolymers. It was also observed that  $G'$  become higher than  $G''$  at low frequencies indicating that a refined morphology was obtained. A significant improvement in compatibilisation and toughness of recycled PS and SAN was achieved through the use of SEBS. The evidence of compatibilisation was obtained from the impact properties and the mechanical properties. The result shows that adding SEBS to rPS/SAN blends improves toughness and elongation but losing strength and stiffness because of its rubbery character. The SEBS acts as an interfacial agent between the matrix and the dispersed phases. It lowers the interfacial tension and improves interfacial adhesion. These results were confirmed by SEM observations which revealed that the incorporation of SEBS was effective in reducing the domain size of the recycled PS or SAN dispersed phases. The DSC analysis and the SEM photos confirmed that SEBS could greatly enhance the compatibility of the rPS/SAN blends, the overall variations in all properties suggest that the SEBS copolymer could be used as an effective compatibilizer for the rPS/SAN mixtures.

## Acknowledgements

K.B. would like to thank Dr Maria del Pilar Posadas Bernal, Dr Patricia SampedroTejedor and Grupo de Elastomeros for their help in hosting him as a doctoral student to carry out the experimental part of his PhD thesis at the Elastomeros laboratory, Instituto de Ciencia y Tecnologia de Polimeros (ICTP-CSIC) Madrid, Spain. This research was funded by Spanish Ministry of Science and Innovation (MINECO) through the research projects RTI2018-096636-J-100 and PID2020-119047RB-100.

## Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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