

Article

Influence of the Presence of Poly(butylene succinate) in the Poly(ethylene terephthalate) Recycling Process

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Abstract: Poly(butylene succinate) (PBS) is one of the contaminants in the Poly(ethylene terephthalate) (PET) recycling process. It is known that high contents of PBS in PET significantly reduce PET properties, but the effect of low contents on PET has yet to be studied. This work studied the influence of low contents of PBS on recycled PET. Five formulations of PBS in PET were prepared, and the properties of relative affinity, mechanical, thermal, and disintegration under composting conditions were assessed. The solubility parameter indicated that PET and PBS are miscible. However, FESEM images show slight marks of immiscibility, and the mechanical characterization results showed that PBS, even in low contents, reduced the mechanical properties of recycled PET, which proves that the materials are not miscible in the studied contents. The DSC results indicated that PBS could not be quickly detected in PET. However, its presence can be inferred by the reduction in PET crystallization degree. Finally, the presence of PBS up to 15 wt.% does neither affect the disintegration under composting conditions nor the thermal stability of recycled PET. The drop in mechanical properties shows that PBS must be removed from the PET waste stream to preserve the quality of the material.

Keywords: Poly(butylene succinate); Poly(ethylene terephthalate); recycling; contaminant; degradation



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1. Introduction

Poly(ethylene terephthalate) (PET) has become the favorite packaging material for water and soft drink bottles worldwide due to its excellent mechanical properties, chemical resistance, thermal stability, and low production cost [1,2]. Additionally, PET displays good barrier properties towards moisture and water, clarity, and very low weight compared to other bottle materials such as glass [1,3]. PET is currently used in carbonated soft drink bottles such as mineral water, energy drinks, beer, wine, and juices [3–5].

PET production has risen considerably in the last few decades. Of the 390 Mt of plastics produced in 2021, 6% (24 Mt) corresponds to PET [6]. Although PET is used in various applications such as medicine or electronic components, its main application is packaging, its products generally being for single use only. However, PET has received considerable attention for recycling, because unlike other plastics or materials, it is fully recyclable [2,4]. PET from bottles is considered the ideal input material for the recycling process because of its inert character and because it is transparent and has no harmful additives [3,4]. PET is recycled via mechanical recycling, which involves processing in shredding, grinding, and melting. Mechanical recycling is simple and cheaper compared to chemical recycling [2]. However, PET is recycled via an open-loop recycling process in which mixtures can be complex and problems of chemical incompatibility and physical inhomogeneity of the different plastics can arise. For instance, a small concentration of PVC in PET embrittles its structure and turns it yellow [7]. Therefore, it is still necessary to separate post-consumer contaminants from the PET waste stream, which are often biological remains or mixtures

of various substances [1]. Among the contaminants, residues of synthetic plastics and biodegradable polymers can be found.

Biodegradable polymers are a range of materials that are susceptible to conversion to biomass, CO₂, and water by a thermochemical process within a specific time frame and disposal environment [8]. Biodegradable polymers have gained attention for single-use plastics applications such as packaging to reduce the environmental pollution due to plastic waste accumulation and reduce the potential hazard if they reach landfills, soils, or fluvial effluents [9]. However, the efficacy of these polymers in reducing plastic waste accumulation depends on the knowledge and understanding of their biodegradability and degradation and their required disposal environment [9]. Consumers usually have low information about the post-consumption treatment of biodegradable plastics; therefore, they usually discard them among traditional thermoplastics [10]. The residues of biodegradable plastics act as impurities in the recycled materials and could affect their mechanical and structural properties, compromising the quality of the final product [11–13]. Moreover, the separation of these impurities in the process can be complex and expensive.

Some of the biodegradable polymers often used are biopolyesters such as polyhydroxyalkanoates (PHAs) and polylactides (PLAs) and aliphatic polyesters such as poly(ϵ -caprolactone) (PCL), Poly(butylene succinate) (PBS), poly(butylene succinate-co-butylene adipate) (PBSA), and polybutylene(adipate-co-terephthalate) (PBAT). PBS is a biodegradable thermoplastic polymer, also known as Poly(tetramethylene succinate), that belongs to the polyester family [14]. PBS can be obtained by the polycondensation of succinic acid or dimethyl succinate and 1,4-butanediol. Both monomers can be derived from bio- or fossil-based sources [15]. PBS can naturally degrade and has a wide processing window, which makes it suitable for diverse processing techniques such as extrusion, film blowing, injection molding, fiber spinning, and thermoforming. According to European plastics, the global bioplastics production was 2.41 Mt in 2021, of which 3.5% corresponded to PBS, and its production is expected to continue to grow in the coming years [16]. PBS is used by the industry to produce mulching films, biodegradable bags, microfiber sheets and costumes, catering items, and foaming agents; moreover, its applicability is increasing in the automotive industry and medical devices [17,18]. Therefore, PBS has great potential in replacing conventional plastics. However, PBS is expensive, has a limited Young's modulus, and has a susceptibility to sudden degradation during melt processing, especially at high temperatures; thus, it is not attractive to use under high-temperature conditions [15,19]. Due to the presence of this material in various applications, PBS can become mixed with recyclable PET, especially in urban trash containers, which can lead to upcoming interferences in the recycling process.

In previous works, the influence of poly(lactic acid) (PLA), thermoplastic starch (TPS), and polyhydroxy butyrate (PHB) in the properties of recycled PET were studied. The results indicated that the contamination of PET with PLA, PHB, or TPS directly affects the mechanical performance of the recycled material [12]. However, the impurities can be quickly identified by quality control departments using easily accessible techniques such as differential scanning calorimetry (DSC) [20]. Moreover, the literature shows that an increasing content of PBS in PET-PBS (10, 20, and 30 wt.%) reduces the miscibility between PBS with PET; PBS also reduces the thermal stability of the blend and reduces the mechanical properties of PET in similar proportions for all PBS contents. The Young's modulus was reduced by 19%, tensile strength by 47%, and elongation at break in 10% [5]. Still, the effect of small quantities of PBS over a PET matrix has not been studied. Therefore, the present study aims to examine the influence of small quantities of PBS on recycled PET properties.

2. Materials and Methods

2.1. Materials and Sample Preparation

Recycled PET was supplied by Extremadura Torrepet S.L. (Badajoz, Spain), and PBS Bionolle 1020MD was supplied by Showa Denko Europe (Munich, Germany). All materials were dried at 60 °C for 24 h in an air circulation oven. The formulation of PBS in PET in concentrations of 2.5, 5, 7.5, 10, and 15 wt.% were prepared and labeled PET-2.5PBS, PET-5PBS, PET-7.5PBS, PET-10PBS, and PET-15PBS, respectively. The formulations were blended by melt extrusion in a twin-screw extruder (Dupra S.L., Castalla, Spain) with a temperature profile of 200/220/240/260/270 °C from hopper to die and a screw speed of 50 rpm. To obtain standard specimen samples for the mechanical characterization, the formulations were then processed by injection molding in a Sprinter 11 t injection machine (Erinca S. L., Barcelona, Spain) with a temperature profile of 240/260/270 °C.

2.2. Miscibility between the Components and Microstructural Characterization

The relative affinity of two polymers can be estimated using the solubility parameters (δ). Thus, the proximity of both polymer parameters is used to evaluate the miscibility between them. To consider the components of the mixture compatible, the solubility parameters of the polymers should be of the same order. Immiscibility may cause the loss of the mechanical properties and even superficial lamination of the material [11]. The relative affinity of PET with PBS was determined using the Small method [21]. According to the Small method, the relative solubility (δ) can be obtained with Equation (1), which considers the contribution of each molecular group to the overall molecular structure.

$$\delta = \frac{\rho \sum F_j}{M_n}, \quad (1)$$

where ρ is the polymer density, M_n is the molar mass of the repetitive unit, and $\sum F_j$ is the summation of the contributions of all molecular groups of the polymer [21]. The molar attraction constants (F) for several groups are listed in Table 1.

Table 1. Small's molar attraction constants for some functional groups [22].

Group	F (cal ^{1/2} cm ^{3/2} /mol)
-CH ₃	214
-CH ₂ -	133
-CH<	28
>C<	-93
-OH	83
-O-	70
-H (variable)	80–100
>C=O	275

The affinity between PET and PBS was also analyzed with a microstructure characterization carried out by a field emission microscope (FESEM). The microstructure of the cross-section of the specimens after the tensile test of recycled PET and PET-PBS formulations was analyzed by FESEM ZEISS ULTRA 55 from Oxford Instruments (Oxfordshire, UK) operated at 2 kV. The samples were previously coated with a gold-palladium alloy to make their surface conductive on the Sputter Mod Coater Emitech SC7620, Quorum Technologies (East Sussex, UK).

2.3. Mechanical Characterization

The mechanical properties of the formulations were assessed in terms of the tensile parameters, hardness, and impact strength. The tensile properties of the formulations were determined in a universal test machine, Ibertest Elib30 of SAE Ibertest (Madrid, Spain), at room temperature, according to the ISO 527 standard [23], at a speed of 10 mm/min and with a load cell of 5 kN. Five specimens of each formulation were tested. The mean value and the standard deviation of the tensile strength, Young's Modulus, and elongations at break are reported.

Hardness tests were performed in a Shore durometer Model 673-D from Instrument J.Bot, S.A. (Barcelona, Spain) using the D scale according to the ISO 868 standard [24]. Samples with 4 mm thickness were tested. The mean value and the standard deviation of 20 measurements are reported.

The resistance to the Charpy Impact was measured in a Metrotec S.A. machine (San Sebastian, Spain), using a 1 J pendulum under the ISO 179 standard. At least three specimens were tested, and the mean was reported.

2.4. Thermal Characterization

Thermal properties were determined by differential scanning calorimetry (DSC) performed in a Mettler Toledo 821 calorimeter (Mettler Toledo, Schwerzenbach, Switzerland). The thermal cycle consists of the first heating from 30 to 280 °C, followed by cooling from 280 to 30 °C, and a second heating from 30 to 350 °C. The analyses were conducted in a nitrogen atmosphere (66 mL/min). The heating and cooling rates in all the cycles were 10 K/min. The percentage of crystallinity (X_c) was calculated with Equation (2).

$$\%X_c = \frac{\Delta H_m}{\Delta H_f \times (1 - w)} \times 100, \quad (2)$$

where ΔH_m is the melting enthalpy of the material (J/g), ΔH_f is the melting enthalpy of 100% crystalline PET (140.1 J/g), and w is the weight fraction of PBS in the formulation [25]. All measurements were done in triplicate.

2.5. Disintegration under Composting Conditions

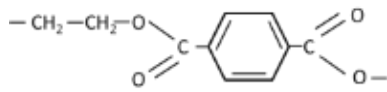
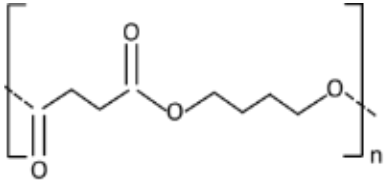
Since this test aimed to verify the influence of PBS in the degradability under the composting conditions of the PET, the test was performed only on the PET-15PBS sample, since this formulation contained the higher amount of PBS among the studied formulations. The PET-15PBS formulation was subject to disintegration under composting conditions at a laboratory-scale level according to the ISO 20200 standard for thermophilic degradation [26]. The dry solid residue was prepared by combining 10% commercial compost (Mantillo, Spain), 30% rabbit food, 10% starch, 5% sugar, 1% urea, 4% corn oil, and 40% sawdust. Then, water was added to the mixture to adjust the final water content to 55%. The wet residue was placed in plastic containers, and the PET-15PBS samples were buried inside it and subjected to aerobic degradation. Before introducing the samples to the container, they were placed in a textile mesh to facilitate the sample collection and to allow access to moisture and microorganisms [27]. The test was carried out for 30 days at a controlled temperature of 58 °C in an air oven. A period of 30 days is considered a reasonable time for the material to go from landfill to recycling. During the test, the solid residue was regularly mixed, and water was added to guarantee aerobic conditions and relative humidity [28]. The samples were recovered on days 8, 21, and 30 to evaluate their visual appearance and to determine their degree of disintegration by gravimetry. After exposure to composting conditions for 30 days, a thermogravimetric analysis (TGA) was carried out on the PET-15PBS sample using a TGA PT1000 from Linseis (Selb, Germany). The test was performed to determine the effect of the degradation on the thermal stability of the material. The samples' weights were between 15 and 20 mg, and they were heated from 30 °C to 600 °C at a rate of 20 K/min in an oxygen atmosphere.

3. Results and Discussion

3.1. Miscibility between PET and PBS and Microstructural Characterization

Table 2 presents PET and PBS solubility. The theoretical solubility was estimated by the relative affinity of the polymers using Small's method; the calculated solubility values were similar to the data available on polymerdatabase.com for the polymers studied [29]. According to the solubility parameter (δ), PET and PBS have a very close affinity, which indicates good miscibility or at least partial miscibility. However, according to the chemical nature of the polymers, the miscibility or immiscibility also depends on the percentages of each component in the blend sample.

Table 2. Relative solubility of PET and PBS.

Polymer	Structure	$\delta(\text{MPa}^{1/2})$ Calculated	$\delta(\text{MPa}^{1/2})$ Available Data [29]
PET		24.3	17.8–24.8
PBS		22.3	20.9

The surfaces of the samples were analyzed by FESEM to corroborate or discuss the previous results. Figure 1 shows FESEM micrographs of the cross-section of the test specimens after the tensile tests of PET (Figure 1a), PET-5PBS (Figure 1b), and PET-15PBS (Figure 1c). The PET cross-section surface shows ductile behavior with a homogeneous, uniform, and soft surface, and it clearly distinguished the presence of one phase. In comparison, the PET-PBS formulations showed a homogeneous smooth surface with lesser softness than that of recycled PET. The softness reduction could indicate embrittlement of the structure due to the presence of PBS. Some irregularities were observed in the PET-5PBS (Figure 1b) and PET-15PBS (Figure 1c) images, which could signal superficial delamination, indicating immiscibility or partial miscibility between PET and PBS. The immiscibility can be attributed to the hydrophobic character of PBS and the relative hydrophilicity of PET [25]. However, up to 15 wt.% of PBS in PET, it is impossible to differentiate drastic signs of immiscibility using FESEM.

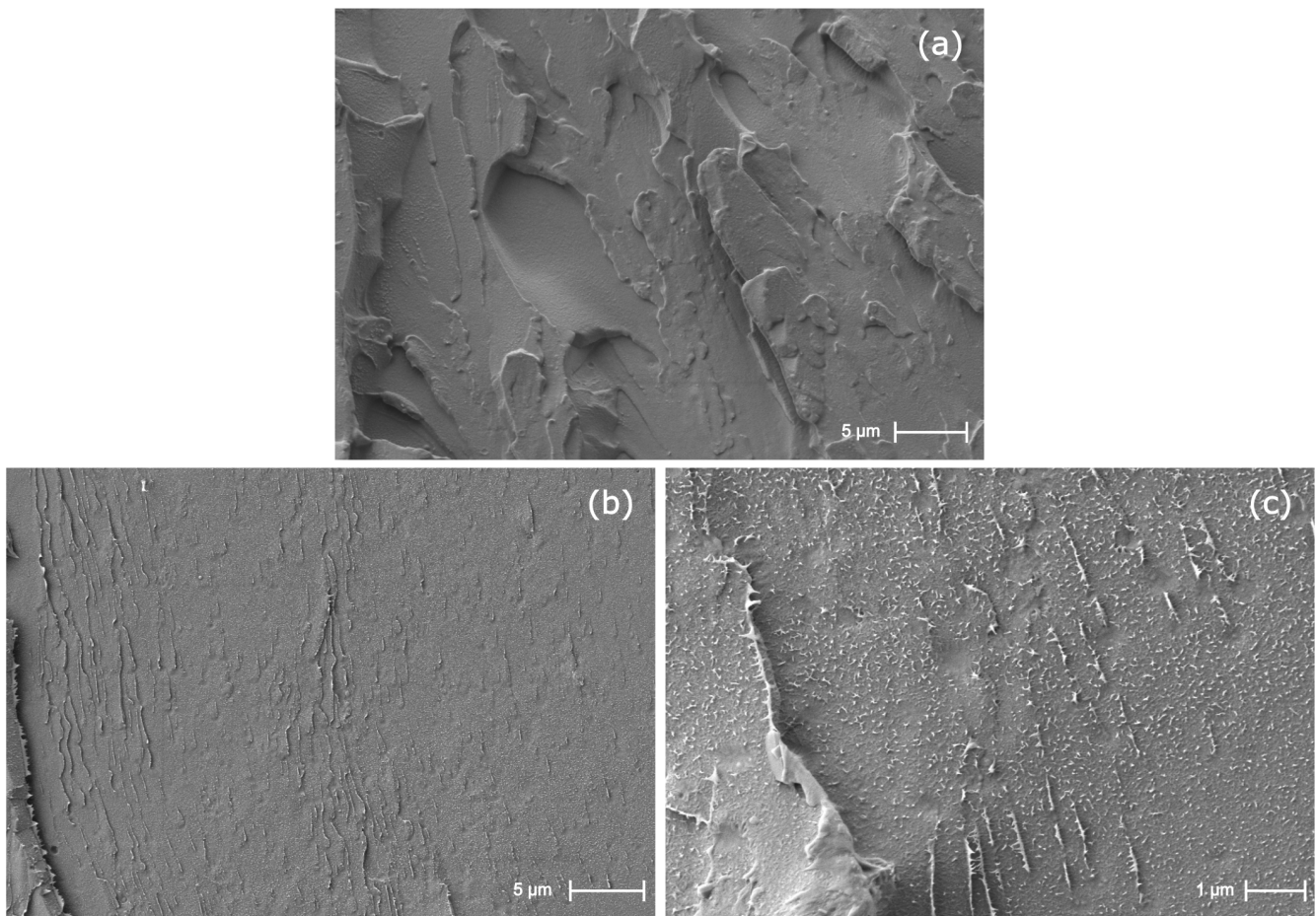


Figure 1. FESEM images of the cross-section of the test specimens after the tensile test of (a) PET, (b) PET-5PBS, and (c) PET-15PBS.

3.2. Mechanical Characterization

Figure 2 shows the mechanical characterization results of the PET-PBS formulations. Recycled PET presents a tensile strength of 62 MPa and Young's modulus of 1683 MPa and an elongation break of 280 MPa, while neat PBS presents a tensile strength of 27 MPa, Young's modulus of 299 MPa, and an elongation break of 198%. In general, the properties of PET decrease when PBS is added to the blend, disregarding the content. The presence of PBS in PET, in contents of 2.5 and 5 wt.%, reduces the PET tensile strength by 33 and 51%, respectively, and PET Young's modulus by 63 and 74%. When the PBS content increases to values of 7.5 wt.% and higher, the tensile strength and Young's modulus of the formulations decrease by 90% concerning the value of recycled PET. Moreover, the PET elongation break drops severely for any PBS content. A significant decrease of the tensile properties of recycled PET, even at the minimum PBS studied content (2.5 wt.%), was determined, pointing to a lack of miscibility in the system. The results highlight the incompatibility between two polymeric matrices and indicate that PBS contamination in the recycling process of PET could be catastrophic, directly affecting the mechanical properties of the recycled material.

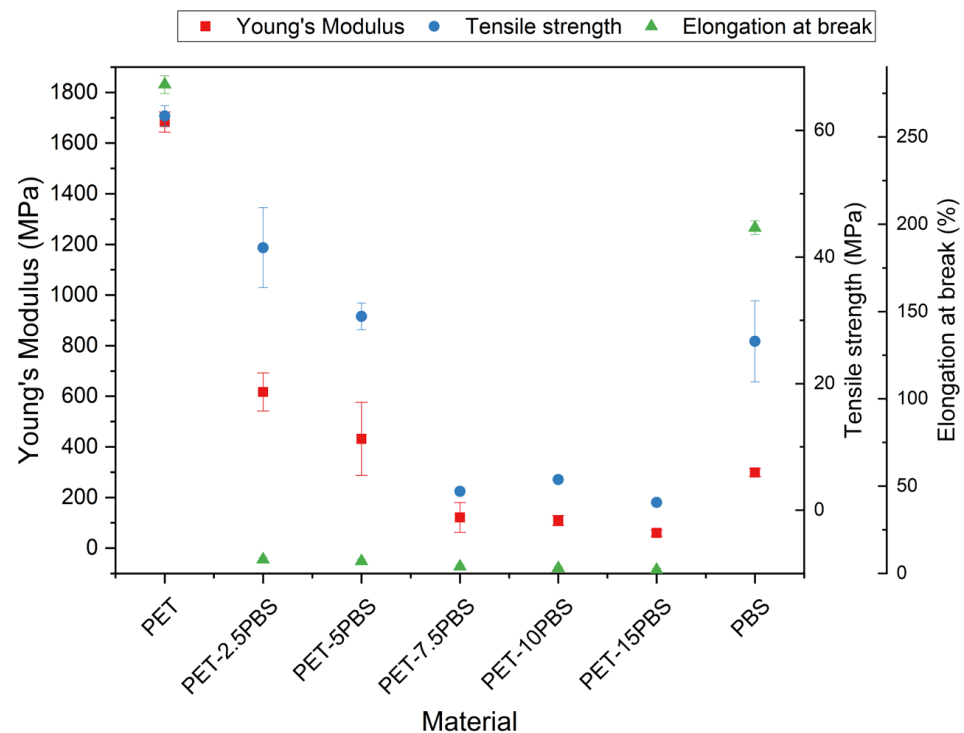


Figure 2. Scatter graphic Young's modulus, tensile strength, and shore D hardness of the PET-PBS formulations.

Regarding other mechanical properties, Table 3 summarizes the impact strength and hardness results obtained for the studied blends. The PET impact strength is reduced by 37% and 47%, adding 2.5 and 5 wt.% of PBS, respectively. At a PBS concentration of 7.5 wt.% and above, the impact strength decreases by 90%. The impact strength results show a loss in the cohesion of the PET structure due to the contamination of PBS in all contents. The recycled PET hardness slightly decreased for all the PET-PBS formulations, with a maximum variation of 5%. The decrease in PET hardness was not influenced by the content of PBS in the system, as seen in the values of the PET-PBS formulations.

Table 3. Mechanical properties of the PET-PBS formulations.

Sample	Impact Strength (kJ/m ²)	Hardness (Shore D)
PET	19 ± 2	81 ± 1
PET-2.5PBS	12 ± 1	76 ± 1
PET-5PBS	10 ± 1	77 ± 1
PET-7.5PBS	2 ± 1	78 ± 1
PET-10PBS	2 ± 1	78 ± 1
PET-15PBS	2 ± 1	79 ± 1
PBS	11 ± 2	63 ± 1

The poor mechanical performance observed in the PET-PBS formulation with PBS contents of 2.5, 5, 7.5, 10, and 15 wt.% indicates that PBS is immiscible with PET, even at low contents [30]. The theoretically calculated solubility using Small's method could indicate that the affinity or partial miscibility between PET and PBS could be obtained only at even lower PBS contents than in this study.

The general drop in the mechanical properties corroborates that PBS must be removed from the PET waste stream to guarantee the properties of the recycled PET.

3.3. Thermal Characterization

The thermal properties determined by DSC are summarized in Table 4, and the DSC curves are shown in Figure 3. PBS presents its melting peak at 112 °C, whereas PET presents its melting temperature peak at 250 °C and its cold crystallization peak at 126 °C, which is observed in the first heating curve. All PET-PBS formulations present a single melting peak, and no T_g is observed. The presence of PBS, in contents of 2.5 to 5 wt.%, do not affect the melting temperature of PET. At contents of 7.5 wt.% and above, PBS impurities cause a slight decrease in the melting temperatures of PET. Additionally, the crystallization degree (χ_c) of PET is diminished by 17% when PBS impurities are present, suggesting that, at any content, PBS contamination interferes with the crystallization of PET during its processing. The slight changes in the DSC curve at the melting peak suggest that identifying PBS impurities quickly with techniques easily accessible to quality control departments such as DSC is complicated in PET contaminated by PBS. DSC was proven to be a feasible technique to detect biodegradable polymers (PLA, PHB, or TPS) as contaminants in PET, and notorious differences were observed [20]. However, the PBS impurities can be identified by a reduction in the crystallization degree of PET.

Table 4. DSC results of the PET-PBS formulations.

Sample	PBS Weight Content (%)	T_g (°C) **	T_{cc} (°C) **	T_m (°C)	ΔH_m (J g ⁻¹)	χ_c (%)
PET	0	72.4 ± 0.2	125.7 ± 0.3	250.1 ± 0.2	-36.6 ± 1.0	26.1 ± 1.0
PET-2.5PBS	2.5	-	-	250.6 ± 0.5	-22.2 ± 1.3	16.3 ± 1.0
PET-5PBS	5	-	-	250.7 ± 0.4	-22.8 ± 1.0	17.1 ± 1.0
PET-7.5PBS	7.5	-	-	247.1 ± 0.8	-23.1 ± 1.6	17.8 ± 1.0
PET-10PBS	10	-	-	247.0 ± 1.0	-20.9 ± 1.5	16.6 ± 1.0
PET-15PBS	15	-	-	245.4 ± 1.3	-20.8 ± 1.2	17.5 ± 1.0
PBS	100	-44 *	-	112 ± 0.2	-42.4 ± 1.0	30.3 ± 1.0

* T_g of PBS was obtained from the literature [31]. ** The T_g could only be determined by DSC in pure PET and pure PBS, while T_{cc} was only in pure PET.

3.4. Disintegration under Composting Conditions

Figure 4 shows the visual appearances of PET-15PBS samples recovered after different days of exposure from the disintegration under composting conditions test. The PET-15PBS sample shows no sign of either biodeterioration, such as a yellowish color or loss of roughness, or degradation, such as fragmentation [32,33]. Figure 5 shows that the weight loss the sample suffered after exposure to the composting conditions is 0.3%, which is negligible. The results reflect the absence of the biodegradation of PET, as previously reported in the literature. Janczak, K. et al. (2018) reported no significant weight loss in PET after six months under greenhouse conditions [1]. Beltrán-Sanahuja, A. et al. [34] reported a PET weight loss of less than 2% in one year in a soil medium. The lack of PET biodegradation under natural environmental conditions is attributed to its aromatic groups, which makes PET resistant to hydrolysis and microorganism degradation, preventing the breakdown of the ester bonds [35]. Moreover, it is seen that the degradation of PBS does not influence the weight loss of the PET-PBS blend, which can be attributed to the composting time, as PBS degradation after 30 days of composting is reported to be only 5% of its total weight [19].

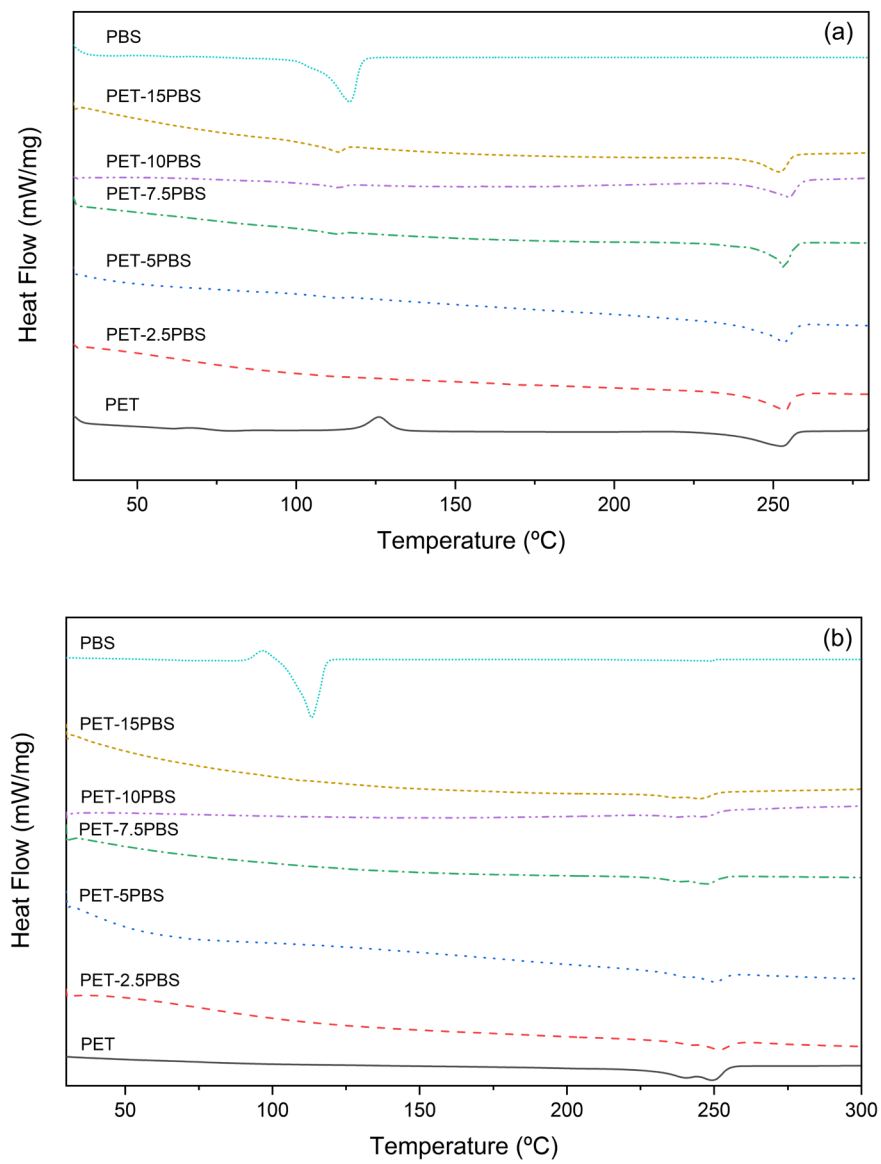


Figure 3. Calorimetric curves of the PET-PBS formulations: (a) first heating and (b) second heating.

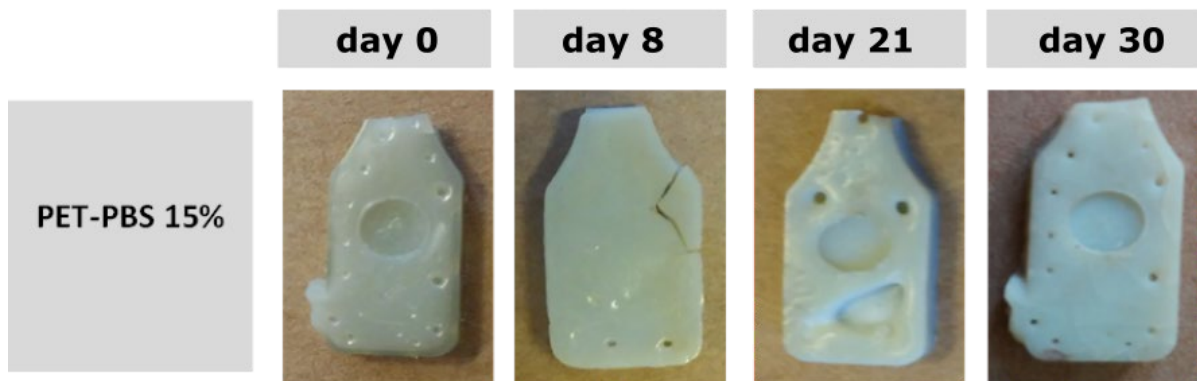


Figure 4. The disintegration of the recycled PET blended with 15% PBS under composting conditions.

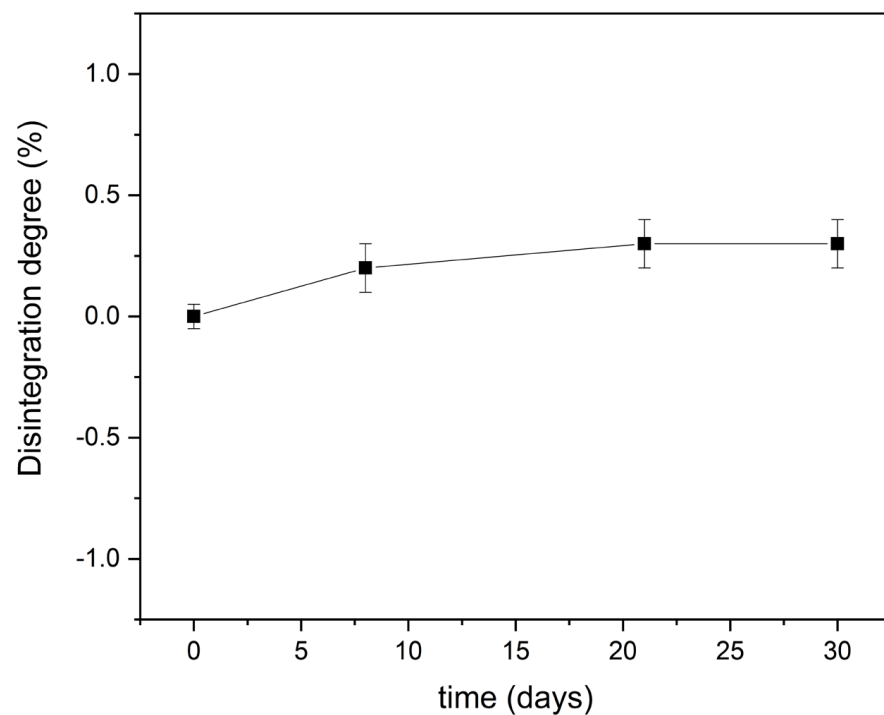


Figure 5. Disintegration degree of the PET-15PBS sample at 8, 21, and 30 days under composting conditions.

Figure 6 shows no significant changes in the sample thermal stability before and after exposure to the composting conditions. It is observed that $T_{5\%}$ before exposure in the composting reactor is 403 °C and T_{max} is 446 °C, with a mass loss of 81.15%.

After composting for 30 days, $T_{5\%}$ is 400 °C and T_{max} is 445 °C, with a loss of mass of 83.14%. The temperatures of PET presented a slight decrease after composting; however, the changes were negligible. The results show that exposure to the composting conditions of the PET-PBS mixture does not generate a diminution in the molecular weight of the PET, as its thermal stability remains unchanged [36]. Therefore, the addition of PBS does not affect the biodegradation of PET in 30 days, which suggests that the material can be further recycled without affecting the degradation of PET.

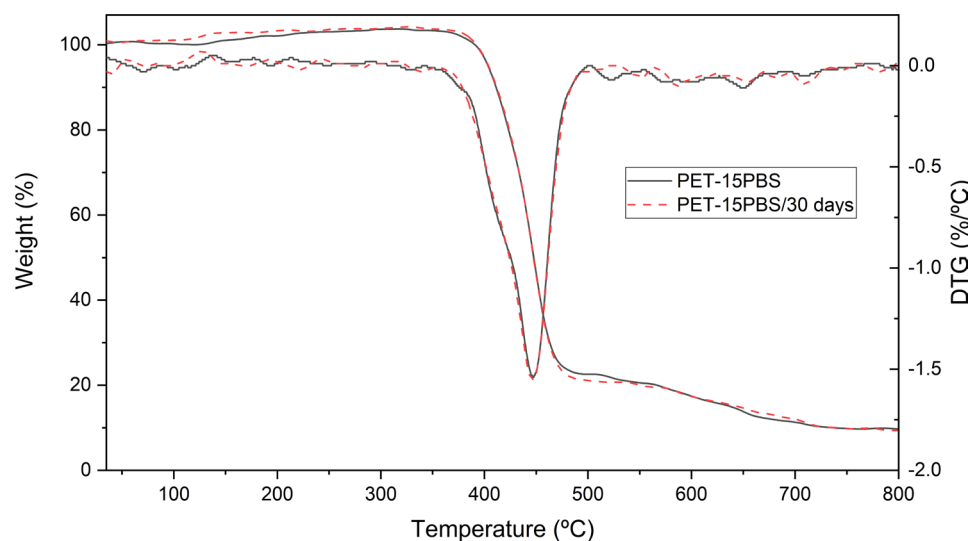


Figure 6. TGA curve of the PET-15PBS formulation before and after 30 days under the compost conditions.

4. Conclusions

The influence of the presence of Poly(butylene succinate) (PBS) as contamination in the recycled Poly(ethylene terephthalate) (PET) matrix was studied. Theoretically, the solubility parameters of PET and PBS show good miscibility between both components. FESEM images of the cross-section of the test specimens after the tensile test show one homogenous phase with no separation for the PET-PBS formulations; however, embrittlement of the structure and slight signs of incompatibility were detected with the addition of PBS, which suggests immiscibility or partial miscibility of the materials for the studied contents. The tensile property results determine that the presence of PBS impurities in recycled PET, in any concentration, causes a significant decrease in the tensile strength, Young's modulus, and elongation break. A decrease in PET impacts the strength and hardness due to PBS contamination was also determined. The DSC analysis revealed that the presence of PBS impurities reduces the crystallization degree of recycled PET by 17%. Moreover, PBS in contents of 7.5 wt.% and above reduces the melting temperature of recycled PET at 3 to 5 °C. The slight changes in the DSC curve at the melting peak make it hard to detect PBS contamination in PET with techniques easily accessible to the quality control department. However, the reduction in the crystallization degree of PET can signal the presence of PBS in the material. The disintegration under the composting conditions analysis determined that the PET-15PBS formulation does not present an important degradation after 30 days of testing, presenting a mass loss of less than 1%. In addition, TGA shows no important change during the thermal degradation of samples before and after exposure to the composting conditions. The degradation and TGA results indicate that PBS impurities do not affect either the thermal stability or the degradation under the composting conditions of PET. The reduction in the mechanical performance of recycled PET due to the contamination of PBS and the difficulty to quickly detect the impurities by control quality techniques such as DSC demonstrates that this biodegradable polymer must be removed from the PET waste stream to preserve the quality of the recycled material.

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Conflicts of Interest: The authors declare no conflict of interest.

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