

# Poly (Butylene Succinate) and PBS Copolyesters

## Degradation

Subjects: Polymer Science  
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The impact of plastics on the environment can be mitigated by employing biobased and/or biodegradable materials (i.e., bioplastics) instead of the traditional “commodities”. Poly (butylene succinate) (PBS) emerges as one of the most promising alternatives due to its good mechanical, thermal, and barrier properties, making it suitable for use in a wide range of applications. Nevertheless, less information regarding PBS biodegradation is available, as research is still ongoing. PBS degradation methods include hydrolytic degradation, enzymatic degradation, and biodegradation in environmental conditions, such as burial, activated sludge, and compost.

Keywords: Degradation ; Poly (butylene succinate) ; Hydrolytic Degradation ; Enzymatic Degradation ; Environmental Degradation

### 1. Hydrolytic Degradation

One of the most common mechanisms of polymer degradation is hydrolytic degradation. In this case (and in the case of enzymatic degradation), the degradation rate depends on poly (butylene succinate) (PBS) crystallinity. Hydrolytic degradation occurs faster in the lower density amorphous regions, facilitating water penetration. This phenomenon causes an increase in the overall degree of crystallinity due to the faster degradation of amorphous domains (that can crystallize once degraded) compared to the more crystalline ones <sup>[1]</sup>. Some researchers report no variation in weight for PBS when exposed to hydrolytic degradation <sup>[2]</sup>, while others report low weight loss <sup>[3][4]</sup>. One study reported a ~31% weight loss for PBS after 24 weeks of hydrolytic degradation at 37 °C. This result could be explained due to the relatively low crystallinity (~56%, as determined by DSC) of the PBS used <sup>[5]</sup>. The pH of the media is also an important parameter that must be taken into account. Morales-Huerta et al. reported a 10% weight loss for hydrolytic degradation at pH = 7.4 after 30 days, whereas the weight loss increased to values higher than 25% for a pH = 2.0 media <sup>[6]</sup>.

As can be deduced from different studies, PBS can be effectively degraded by the hydrolysis of the ester bonds, achieving different results depending on many different parameters involved, such as the synthesis method, MW, crystallinity, or the experimental conditions of the biodegradation assays.

### 2. Enzymatic Degradation

So far, enzymatic degradation is regarded as one of the most attractive and effective methods for the biodegradation of biopolyesters. The main reason is the presence of labile ester bonds in the chemical structures of biopolyesters, where enzymes can attack <sup>[7]</sup>. Then, the enzymatic degradation process usually starts with the attachment of the enzyme on the surface, and hydrolysis proceeds via surface erosion. Among all the different types and families of enzymes that can effectively biodegrade PBS and its copolymers. **Table 1** shows various enzymes and different experimental conditions for PBS enzymatic biodegradation.

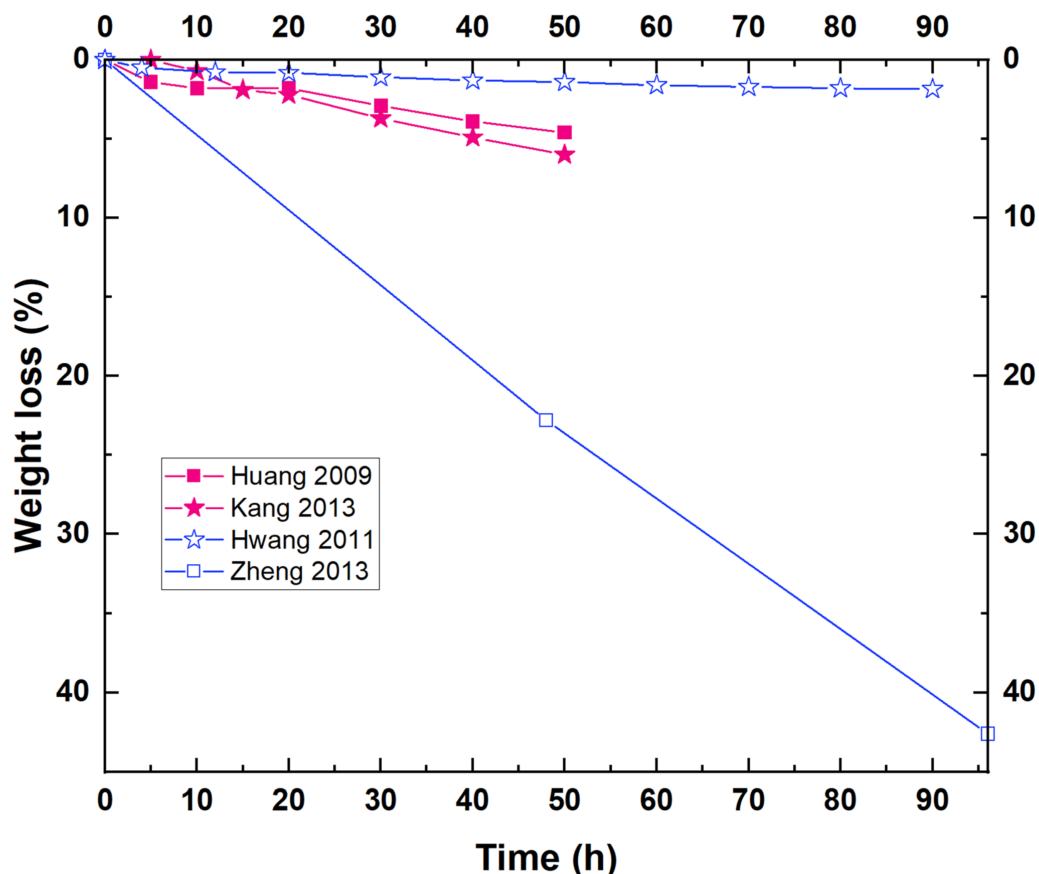
**Table 1.** Classification of enzymes by families for different PBS enzymatic degradation studies.

Family	Enzyme	Substrate	Experimental Conditions	Results	Reference
Cutinase	<i>Fusarium solani</i>	PBS films 30 × 10 × 0.1 mm <sup>3</sup>	pH = 8.0 at 40 °C, 20 µg/mL	100% weight loss in 6 h	[8]
Cutinase	<i>Pichia pastoris</i>	PBS films 30 × 10 × 0.5 mm <sup>3</sup>	pH = 7.4 at 37 °C, 0.15 mg/mL	100% weight loss in 12 h	[9]

Family	Enzyme	Substrate	Experimental Conditions	Results	Reference
Cutinase	<i>Fusarium solani</i>	PBS films 30 × 10 × 0.1 mm <sup>3</sup>	pH = 7.4 at 37 °C, 10 mg/mL	98.4% weight loss in 12 h	[10]
Cutinase	<i>Fusarium solani</i>	PBS films 30 × 10 × 0.5 mm <sup>3</sup>	pH = 7.2 at 37 °C, 18 U/mL	~100% weight loss in 26 h	[11]
Lipase	<i>Candida antarctica</i> (CALB)	PBS films 30 × 10 × 0.5 mm <sup>3</sup>	pH = 7.2 at 45 °C, 18 U/mL	95.1% weight loss in 26 h	[11]
Lipase	<i>Candida rugosa</i>	PBS films 10 × 10 × 0.5 mm <sup>3</sup>	pH = 7.4 at 30 °C, 0.1 mg/mL	2% weight loss after 7 weeks	[12]
Lipase	<i>Pseudomonas cepacia</i>	PBS films 20 × 30 × 0.3 mm <sup>3</sup>	pH = 8.0 at 40 °C, 0.06 mg/mL	2% weight loss after 90 h	[13]
Lipase	<i>Candida antarctica</i> (CALB) N435	PBS films 30 × 10 mm <sup>2</sup>	pH = 7.4 at 37 °C, 1.2 mg/mL	1.8% weight loss after 90 h	[14]
Lipase	Porcine pancreas	PBS films 30 × 10 mm <sup>2</sup>	pH = 7.4 at 37 °C, 0.8 mg/mL	0.9% weight loss after 90 h	[14]
Lipase	<i>Pseudomonas cepacia</i>	PBS films 10 × 10 × 0.1 mm <sup>3</sup>	pH = 6.86 at 45 °C, 0.22 mg/mL	4.6% weight loss after 50 h	[15]
Lipase	Porcine pancreas	PBS film discs 10 × 10 × 0.2 mm <sup>3</sup>	pH = 7.4 at 37 °C, 1 mg/mL	21% weight loss after 30 days	[6]
Lipase	<i>Pseudomonas fluorescens</i>	PBS films 10 × 10 × 0.2 mm <sup>3</sup>	pH = 7.3 at 37 °C, 2 mg/mL	No visible degradation after 300 h	[16]
Lipase	<i>Pseudomonas cepacia</i>	PBS films 10 × 10 × 0.1 mm <sup>3</sup>	pH = 6.86 at 45 °C, 0.53 mg/mL	100% weight loss after 288 h	[17]
Lipase	<i>Pseudomonas cepacia</i>	PBS film discs 20 × 20 × 0.05 mm <sup>3</sup>	pH = 7.4 at 37 °C, 1 mg/mL	6% weight loss after 50 h	[18]
Lipase	<i>Rhizopus delemar</i> and <i>Pseudomonas cepacia</i>	PBS film discs 50 × 50 × 2 mm <sup>3</sup>	pH = 7.2 at 30 °C, 0.09 & 0.01 mg/mL	2% weight loss after 360 h	[19]

Enzymatic degradation assays for biopolyesters and PBS are commonly carried out at physiological temperature (i.e., 37 °C) [6][10][13]. However, it has been demonstrated that this degradation method is favored at a temperature close to  $T_m$  (the PBS melting temperature is above 100 °C) [20]. Some researchers have reported low weight loss values for PBS homopolymer at different experimental conditions, reaching a 3.5% weight loss after 12 days in the presence of *Pseudomonas cepacia* lipase [20], or even lower [12][14]. The low degree of degradation obtained could be attributed to the high crystallinity of this polymer compared to other aliphatic polyesters [20]. Other studies carried out under different experimental conditions report much higher degradation rates. For example, for enzymatic degradation assays employing cutinases, weight losses reach almost 100% in just 12 h [10], as seen in **Table 1**. An interesting study developed by Shi et al. showed the influence of two different enzymes (*Fusarium solani* cutinase and *Candida antarctica* lipase B, CALB) in the degradation rate of PBS. They found that the PBS degradation rate was much faster by the action of cutinase. PBS degraded in the presence of cutinase reached ~50% weight loss in 4 h, whereas those degraded in the presence of lipase reached ~20% weight loss over the same time. For both cases, a nearly total decomposition was achieved after 26 h [11].

**Figure 1** shows the results corresponding to different biodegradation studies where the weight loss (%) of PBS in the presence of lipase from *Pseudomonas cepacia* has been reported. As can be observed in **Figure 1**, only one study shows a relatively high weight loss of PBS with this enzyme (higher than 40% in 100 h) [17]. For the rest, the weight loss reached after several hours in contact with *Pseudomonas cepacia* is very low (less than 6%), which could be attributed, in part, to the low concentration of the enzyme employed for some of the studies [13][15][18].



**Figure 1.** Weight loss curves corresponding to various enzymatic degradation studies of PBS by the action of a lipase from *Pseudomonas cepacia*. Experimental conditions differ from one study to another, although the same enzyme is used.

### 3. PBS-Based Copolymers: Hydrolytic and Enzymatic Degradation

In the case of PBS copolymers, different (and opposite) results are reported depending on the nature of the second comonomer. For instance, the biodegradability of aromatic polyesters is less favored than in the case of aliphatic polyesters such as PBS [6]. Thus, incorporating a second comonomer in the structure of PBS could favor or prevent the degradation of the polyester, attending to the nature of the second constituent.

Hydrolytic and enzymatic degradation of PBS-*ran*-PBFur copolyesters have been determined by placing 60:40 and 40:60 copolymers in a pH = 2.0 or pH = 7.4 medium at 37 °C [6]. Firstly, the enzymatic degradation was more effective than hydrolysis, reaching 15–20% of weight loss versus 3–5% for the latter (hydrolysis) after 30 days. Furthermore, the behavior of the copolyesters was more similar to that of PBS in the case of enzymatic degradation. Regarding the acidic medium, the results were in between enzymatic and hydrolytic degradation, but far away from those obtained for the PBS homopolymer, as the homopolymer achieved a 30% of weight loss, compared to the 10–15% weight loss of the copolymers.

Han et al. studied the enzymatic degradation behavior for different poly(butylene succinate-*ran*-butylene 2-methylsuccinate) (PBS-*ran*-PBMS) copolyesters, reporting higher degradation rates for those copolymers with a higher PBMS content. Considering the copolymer with 20% mol in PBMS, the hydrolytic degradation (without the enzyme) showed a negligible weight loss compared to that of the enzymatic degradation (amano lipase from *Pseudomonas fluorescens*), achieving a 30% weight loss in 300 h [16].

The copolymerization of PBS with salicylic acid was studied as an attempt to produce polymer films with potential applications in agricultural applications. Enzymatic degradation assays carried out in the presence of *Candida antarctica* lipase B (CALB) showed very low degradation after 20 days (~1.5 wt% for neat PBS); however, the addition of salicylic acid increased this value up to ~3.5 wt% [21].

In the case of the enzymatic hydrolysis of PBS and PBS-*ran*-PBA copolymers in the presence of *Candida cylindracea* lipase [22], the highest degradation was obtained for the copolyesters containing 25% and 50% mol of butylene succinate, reaching 20% and 30% of weight loss, respectively, after 90 h. It is necessary to remark that the enzymatic degradation is not affected by the MW; hence, similar results are obtained for low MW (i.e.,  $M_w$  of 6300 g/mol) and high MW (i.e.,  $M_w$  of 29,000 g/mol) [23].

## 4. Biodegradation in Environmental Conditions

Although enzymatic hydrolysis (laboratory conditions) has shown satisfactory results for PBS biodegradation, this biopolyester commonly degrades in environmental conditions [24]. The study of the biodegradation of PBS under environmental conditions will give an idea for the implementation of PBS in agricultural applications such as mulching films [25][26][27]. PE films are commonly employed, being an effective method for promoting plant growth during the cold seasons (i.e., spring and autumn). The problem here is the recyclability of the PE film due to the contamination caused to the soil itself, so a biodegradable film is required, and PBS is a suitable candidate to solve this issue [28].

The experiments for this type of biodegradation are usually carried out following different standards from international organizations (ISO, ASTM, and EU). Because of this, the definition of more experimental parameters is required as compared to enzymatic and hydrolytic assays. As the conditions and parameters differ from one study to another (as well as the soil employed for the tests and the microorganisms content in the soil), biodegradation in environmental conditions covers a wide range of variable results [29].

### 4.1. PBS Homopolymer and PBS-Based Copolymers

PBS biodegradation in environmental conditions usually takes more time as compared to enzymatic/hydrolytic PBS degradation. Kim et al. reported a low degradation of PBS when exposed to environmental degradation (below 8% weight loss after 120 days) [30]. Similar trends have been obtained by Huang et al. (below 3% weight loss in 100 days) [31] and other reports [32]. However, the study of PBS biodegradation in a controlled compost at 58 °C (based on ISO 14855-2) showed that PBS powder biodegradation reached 60% weight loss in 40 days and increased to 80% in less than 80 days. These results are highly promising, opening a path for the establishment of experimental protocols to determine the environmental biodegradation of this aliphatic polyester [33]. Kunioka et al. also reported very high biodegradation rates for PBS in powder form, reaching almost 80% weight loss in less than 80 days [33]. These outstanding results are explained as the PBS was tested in powder form, which differs from the tensile specimens commonly used to determine environmental biodegradation.

The biodegradation of PBS and PBS-*ran*-PBA copolymers subjected to different environments, as biodegradation in compost, soil, and artificial weathering, has been reported. For the artificial weathering, both polymers were submitted to UVA radiation and artificial rain, whereas in soil and compost experiments, no radiation was employed. For the first assay, a ~30% weight loss was achieved for PBS in 24 weeks (~50% in the case of the PBS-*ran*-PBA copolymer). In contrast, biodegradation in soil and artificial weathering showed negligible degradation for PBS, while PBS-*ran*-PBA presented a ~20% weight loss for the biodegradation in soil experiment and negligible for artificial weathering [34]. In another study, the biodegradation of PBS-*ran*-PBFur copolymers in compost at 58 °C showed the best results for the 20 mol% of furanoate composition, achieving an almost 100% of degradation in 80 days [35].

### 4.2. PBS-Based Biocomposites

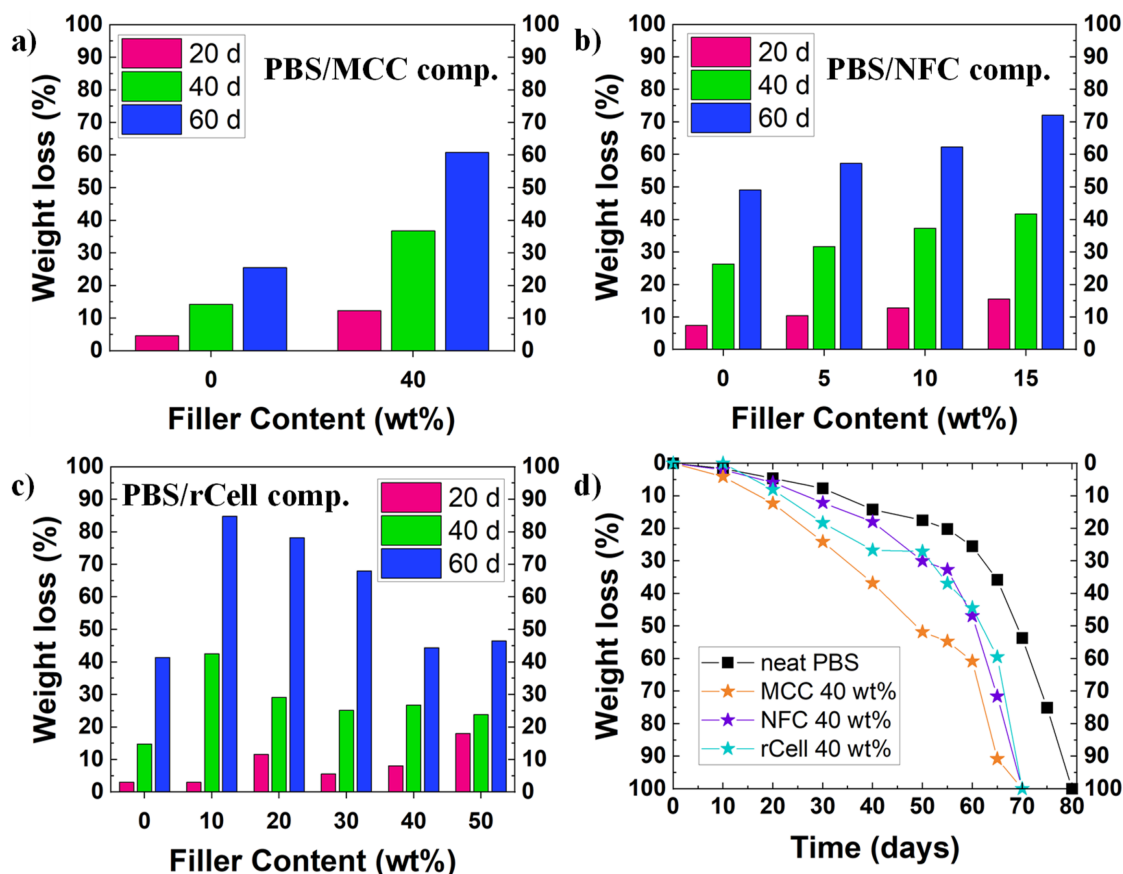
The presence of fillers in PBS biocomposites has been widely studied to modulate the degradation in environmental conditions, as in the case of other types of degradation and thermomechanical and barrier properties. Special cases are natural fillers, that, in addition to being easily biodegraded, can potentially increase the degradation rate of PBS. **Table 2** includes several examples of biodegradation studies carried out under environmental conditions for different PBS-based biocomposites. Among all the examples presented in the table, some interesting results will be commented on below. As a general idea to consider, the trend shows that PBS-based biocomposites degrade faster than neat PBS.

**Table 2.** Different biodegradation studies of PBS and PBS biocomposites carried out in environmental conditions.

Filler	Filler Content	Experimental Conditions	Results (Weight Loss)	Reference
Rubberwood powders (RWP)	0–40 wt%	60 days, no UV radiation, water control each 48 h	<1% (PBS) 2–10% (PBS/RWP)	[32]
Rice husk flour (RHF) and wood flour (WF)	0–40 wt%	4 months	7% (PBS) 8–12% (PBS/RHF and PBS/WF)	[30]
Sugarcane rind fiber (SRF)	0–15 wt%	100 days, natural soil in cropland, water control each 24 h	2.5% (PBS) 10–20% (PBS/SRF)	[31]

Filler	Filler Content	Experimental Conditions	Results (Weight Loss)	Reference
Microcrystalline cellulose (MCC) and nanofibrillated cellulose (NFC)	0–40 wt%	75 days, simulated compost, 58 °C, pH = 5.7–6.3, 50 wt% water content	100% in 75–80 days (PBS) 100% in 65–70 days (PBS/MCC and PBS/NFC)	[36]
Cotton fiber (CF)	0–40 wt%	Based on ISO 14855-2 100 days, 58 °C, 10 mL/min air flow	~60% (PBS) ~90% (PBS/CF)	[37]
Rice husk flour (RHF)	0–40 wt%	Based on ASTM D 6003-96 80 days, 30 °C, pH = 7, 50–60 wt% water content	~12% (PBS) 13–18% (PBS/RHF)	[38]
Jute fiber (JF)	0–30 wt%	180 days, compost soil, 30 °C, constant water control	31.4% (PBS) 47.3–62.5% (PBS/JF)	[39]
Abaca fiber (AF)	10 wt%	180 days, black soil and leaf mold for gardening, 25–30 °C, water control each 48 h	~30% (PBS) ~50% (PBS/AF)	[40]
Soy, canola, and corn gluten meals (SM, CM, CGM) and switchgrass (SG)	25 wt%	Based on ASTM D6400 200 days, 3 month-old compost, 58 °C, pH = 7–8, 50–55 wt% water content	~95% (PBS) ~85% (PBS/SG) 90–95% (PBS/SM, PBS/CM and PBS/CGM)	[41]
Organically modified montmorillonite (OMMT)	0–10 wt%	180 days, natural compost, 30 °C, pH = 7.46, 60–70 wt% water content	~9% (PBS) ~3.5–5% (PBS/OMMT)	[42]
Nanofibrillated cellulose (NFC) and recycled cellulose (rCell)	0–15 wt% (PBS/NFC) 0–50 wt% (PBS/rCell)	80 days, 58 °C, pH = 5.7–6.5, >50 wt% water content	~80% (PBS) ~85–92% (PBS/NFC) 100% in 70 days (PBS/rCell)	[43][44]
Pistachio shell flour (PSF)	0–30 wt%	Based on ISO 20200 112 days, compost, 58 °C, 55 % relative humidity	~18% (PBS) ~14–17.5% (PBS/PSF)	[45]

Although the experimental conditions differ from one study to another, it has been observed that cellulose fillers (micro- or nano-sized) achieve one of the best results for PBS degradation. Platnieks et al. have studied many different PBS/cellulose-based composite films: microcrystalline cellulose (MCC) [36], nanofibrillated cellulose (NFC) [14][23], and recycled cellulose from TetraPak® [44]. The experimental conditions were similar for all the studies, employing a simulated compost under aerobic conditions at 58 °C, with a slightly acidic medium (pH = 5.7–6.5) and a water content of 50% or higher. The researchers found that, although almost every sample was completely disintegrated within 75–80 days, PBS-biocomposites degraded 5–10 days earlier than neat PBS films. In general, the degradation rate was faster at higher filler content. However, for the PBS/rCell biocomposite with the highest content in rCell, the degradation rate was faster in the early stages of the assay, whereas it slowed down during the course of the experiment (see **Figure 2a–c**). If the results were compared to corresponding to the PBS biocomposites with high filler content (i.e., 40 wt%), it was found that the PBS/MCC composite degraded faster than the other two biocomposites (i.e., PBS/NFC and PBS/rCell) which present a similar behavior, being much faster than the biodegradation of neat PBS (see **Figure 2d**).



**Figure 2.** Biodegradation in environmental conditions for several PBS/cellulose-based biocomposites: influence of filler content in weight loss for (a) PBS/MCC composites, (b) PBS/NFC composites, and (c) PBS/rCell composites; (d) variation of biodegradation rates for PBS and the three PBS/cellulose-based biocomposites. In **Figure 2a–c**, weight losses at 20, 40, and 60 days are represented.

Other PBS biocomposites include rubberwood powder (RWP) from sawdust wastes as a natural filler (lignocellulosic nature). The environmental degradation of these PBS/RWP biocomposites was studied, showing a ~10 wt% weight loss after 60 days of soil burial testing. This behavior was attributed to the decrease in the crystallinity of the PBS biocomposites with the increasing content of RWP [32]. In another example, the biodegradation of the PBS biocomposites with rice husk flour (RHF) and wood flour (WF) in soil burial testing showed a 10 wt% degradation for the RHF composites after 120 days [30]. In both cases, the weight loss was directly related to the biocomposite content, increasing with the filler content.

This entry is adapted from [10.3390/polym14051025](https://doi.org/10.3390/polym14051025)

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