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# Investigation of the recyclability and compostability of biopolymers contaminated by petroleum-based polymers

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**Abstract.** Nowadays, we can choose a carrier bag made of traditional LDPE or a biodegradable polymer to pack vegetables, bakery products, and other products in more and more shops. However, the customers and the selective waste collection system are not yet prepared for the separate collection of compostable biopolymers. Therefore, they are mixed in the plastic waste stream. Therefore, the aim of the study was to analyze the mechanical and optical properties, and the compostability of different low-density polyethylene (LDPE) and poly(butylene adipate-co-terephthalate) (PBAT) compounds. We made different compounds from LDPE and PBAT by twin-screw extrusion and blown films from the regranulates. We investigated the tensile and optical properties and the biodegradability of the blown films. The tensile test showed that the "contaminants" had a more significant effect on elongation at break than tensile strength. We observed that the haze of the LDPE-based blends increased with an increasing weight fraction of PBAT. We found that PBAT-based samples were completely disintegrated in 42 days, regardless of the weight fraction of LDPE.

## Introduction

Much of the plastic waste is still deposited or thrown away in the environment. Therefore, environmentally conscious manufacturers in the packaging industry are increasingly using bioplastics besides or instead of petroleum-based polymers. Most petroleum-based polymers (40%) are used in the packaging industry. Similarly, a significant proportion of biodegradable plastics are made into flexible (43%) and (16%) rigid packaging. However, plastic packaging, e.g. single-use plastic bags, have a very short lifetime; therefore, they become waste within a few weeks on average [1-5].

The recycling of petroleum-based polymers is already well established, and there are also several possible ways of recycling biodegradable polymers (e.g. mechanical recycling, home and industrial composting). However, the problem of selective collection and the treatment (recycling and, or composting) of biodegradable plastic waste has not yet been resolved therefore they can mix with petroleum-based polymers in the waste stream [1, 6]. Some publications [7-9] have already analyzed the influence of biodegradable polymer (mainly PLA) "impurity" on the recycling process of petroleum-based polymer waste. Besides, contaminants can be a problem during composting, as the quality of compost deteriorates with non-biodegradable materials (e.g., petroleum-based plastics), and the time and cost of processing waste can increase significantly [10].

Gere and Czigany [7] and Lu *et al.* [8] also observed that injection molded samples of polyethylene and poly(lactic acid) blends became brittle with the appearance of PLA "contaminants," and elongation at break was reduced.

LDPE is the most popular flexible packaging material in the case of petroleum-based polymers and PLA (e.g. clear films, nets) and PBAT (stretch films, labels) in biopolymers [3, 11]. Therefore, the aim of this paper is not to develop a new blend to improve the properties of PBAT or LDPE. The novelty of this manuscript is to investigate the influence of LDPE contamination in the biodegradable plastic (PBAT) waste stream in the recycling and composting process. Furthermore, we also investigated PBAT impurities in LDPE plastic waste.

## Experimental

### Materials

We used LDPE type TIPOLEN FA 244-51 (MFI (190 °C, 2.16 kg): 0.3 g/10 min, density: 0.92 g/cm<sup>3</sup>) supplied by MOL Petrochemicals Co. Ltd. (Hungary) as the petroleum-based component, and PBAT blend type BF 7210 (MFI (190 °C, 2.16 kg): 5.5 g/10 min, density: 1.38 g/cm<sup>3</sup>), supplied by BioComp (USA), as the bio-based component. It is a PBAT-based polymer which contains PLA and CaCO<sub>3</sub>. These types of materials are recommended for film production.

We made PBAT-based blends, where the ratio of PBAT/LDPE were 100/0; 99/1; 97,5/2.5; 95/5; 90/10; 0/100 wt/wt%). These blends were dried before processing. We also made LDPE-based blends, where the ratio of LDPE/PBAT were 100/0; 99/1; 97,5/2.5; 95/5; 90/10; 0/100 wt/wt%). These blends were conditioned before processing.

### Material preparation and processing

Before compounding and film blowing, the PBAT-based materials were dried at 80 °C in a Faithful WGLL-125 BE (China) hot air drying oven for 4 hours, and the LDPE-based materials were conditioned at 23 °C and at a relative humidity of 35% in a desiccator for 72 hours.

The different PBAT-based and LDPE-based compounds (regranulates) were produced with a Labtech Scientific LTE 26-44 (Thailand) co-rotating twin-screw extruder (screw diameter: 26 mm, length/diameter (L/D) ratio: 44). The zone temperatures (from hopper to die) were 160–160–165–165–170–170–175–175–180–180 °C. The screw rotation speed was 30 rpm, and the melt pressure was 30–40 bar.

After that, films were prepared from the regranulates via film blowing using a Labtech Scientific LTE 25-30/C (Thailand) single-screw extruder (screw diameter: 25 mm, L/D ratio: 30), equipped with a Labtech Scientific LF 400 (Thailand) film blowing and take-off unit. The zone temperatures (from hopper to die) were 165–170–175–180–180 °C. The screw rotation speed was 30 rpm. We kept film thickness in the range of 15–30 µm by carefully adjusting the blow-up and take-up ratios.

### Methods

Tensile properties were analyzed with a Zwick Z005 multifunctional material tester (Germany) with an AST Mess & Regeltechnik KAP-TC (Germany) load cell (measuring range 0–5000 N, preload 1 N). The tensile strength and elongation at break of ISO 527-3/2 dumbbell-shaped specimens were measured at a tensile speed of 50 mm/min. We tested at least six samples of each blend, and the average of the results was calculated. The tests were performed at 22 °C and at a relative humidity of 50%.

Optical properties were measured with a BYK Gardner haze-gard dual transparency meter (Germany), according to the ISO 13468 standard.

The biodegradability of films was investigated according to the standard ISO 20200. According to the standard, the synthetic, laboratory-scale compost soil contained (as dry %wt): 40% sawdust, 30% rabbit-feed, 10% ripe compost, 10% corn starch, 5% saccharose, 4% corn seed oil, and 1% urea. 55 wt% of distilled water was added to the mixture. The aerobic degradation was carried out in a Climacell 111 climate chamber at a constant temperature of 58±2 °C for 90 days. During this time, the moisture, mixing, and aeration of the samples was periodically controlled according to ISO 20200. We tested 25 mm × 25 mm pieces from each type of film. After 90 days, we sieved the

compost to separate the remaining polymeric pieces greater than 2 mm, as specified in the standard, to calculate the degree of disintegration (Eq. 1).

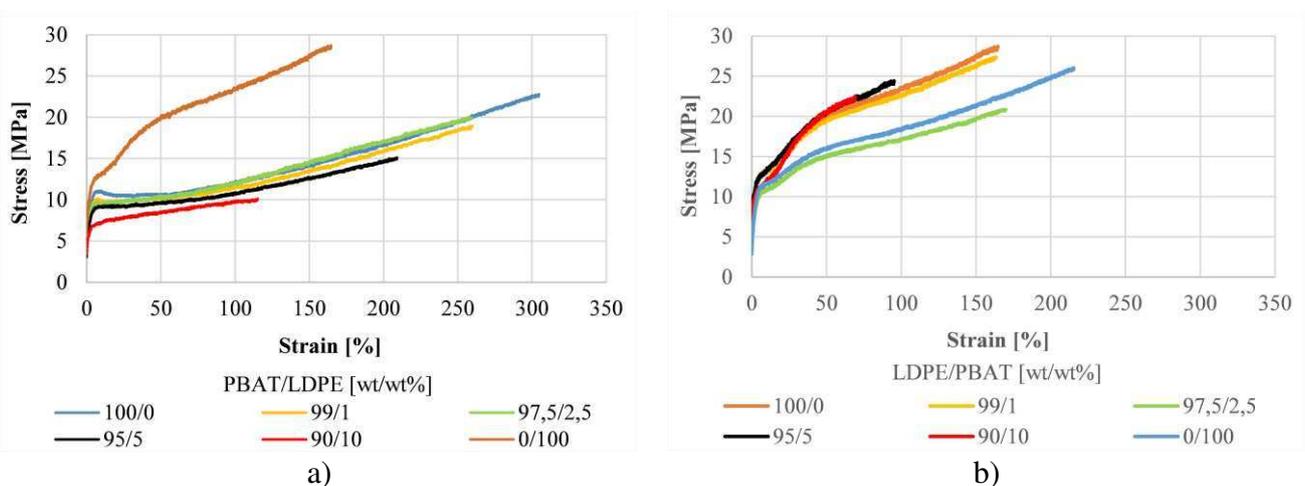
$$D = \frac{m_i - m_r}{m_i} \times 100 \quad (1)$$

where  $D$  is the degree of disintegration,  $m_i$  is the initial dry mass of the test material, and  $m_r$  is the dry mass of the residual test material recovered by sieving.

## Results and discussion

### Mechanical properties of the blends

*Fig. 1* shows the tensile curves of the PBAT-based (*a*) and LDPE-based (*b*) blends of different weight fractions. *Fig. 1a* shows that in the case of PBAT-based blends, pure LDPE was broken at higher stresses but lower elongation at break than PBAT. As the proportion of "contaminants" (LDPE) present in the blend increased, tensile strength and elongation at break also decreased. *Fig. 1b* shows that in the conditioned case (LDPE-based), the elongation at break and tensile strength of PBAT was also significantly reduced compared to the dried samples (PBAT-based). This can be attributed to the hydrolytic degradation caused by the higher moisture content during processing. In the case of LDPE-based blends, the elongation at break of the samples decreased with increasing amounts of PBAT as an impurity. The tensile curves of LDPE were similar in both cases because the properties of hydrophobic LDPE were not affected by drying or conditioning.



*Fig. 1. Tensile curves of PBAT-based (dried before processing) and LDPE-based (conditioned before processing) blends of different weight ratios*

*Fig. 2* shows the tensile strength (*a*) and elongation at break (*b*) of different weight ratios of PBAT-based and LDPE-based blends as a function of PBAT content. 1 wt% "contaminant" in LDPE and 2.5 wt% "contaminant" in PBAT do not affect tensile strength. However, an increase in the amount of contaminant decreased the tensile strength. A similar trend can be observed for elongation at break, but in this case, the contaminants had a more significant effect. This is caused by the fact that LDPE and PBAT are thermodynamically immiscible, therefore there is poor adhesion between the LDPE and the PBAT, and the dispersed particles are present in the blends as defects.

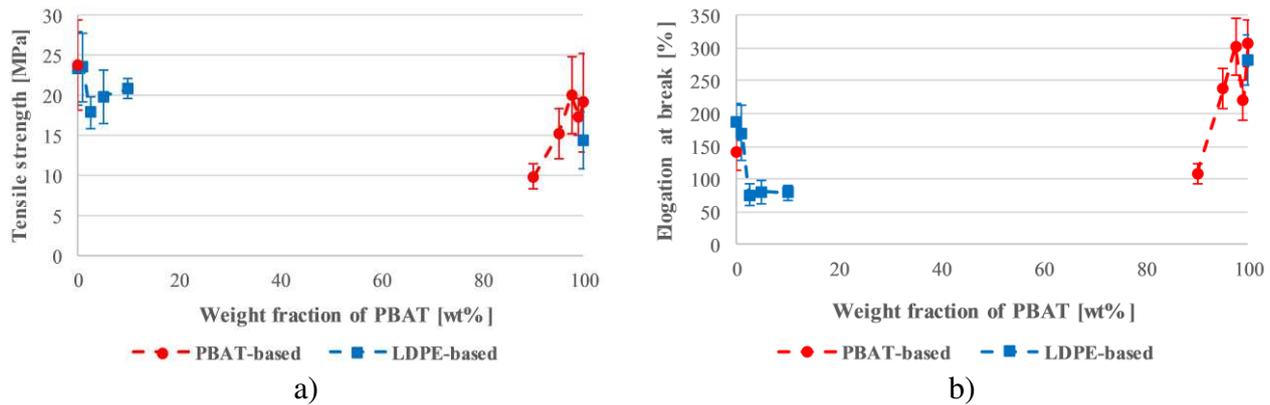


Fig. 2. Tensile strength (a) and elongation at break (b) of different PBAT-based and LDPE-based blends

### Optical properties of the blends

The optical properties of the PBAT-based and LDPE-based blends in different weight ratios are shown in Fig. 3.

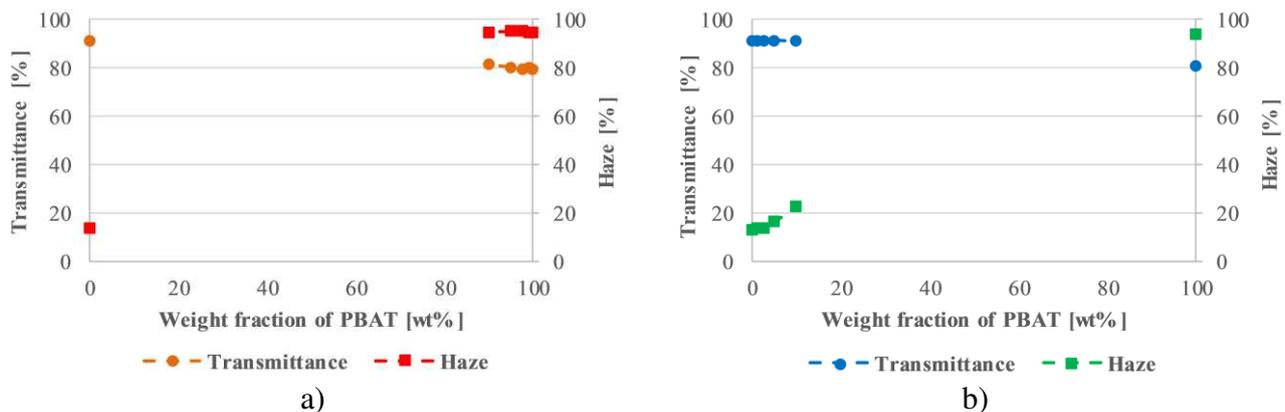


Fig. 3. The optical properties of the different PBAT-based (a) and LDPE-based (b) blends

There was only a small difference in the transmittance of neat LDPE and PBAT films. The transmittance of PBAT films was 10% lower than that of LDPE due to  $\text{CaCO}_3$  mineral filler in PBAT. Drying and conditioning (before processing) did not affect transmittance. It can also be seen that the presence of "contaminants" did not affect the transmittance for any of the blends in the studied range of contaminants content. In contrast, we observed a significant difference in the haze of the neat LDPE and PBAT films. The higher haze is caused by the  $\text{CaCO}_3$  mineral filler and the dispersed PLA phase in the PBAT. Considering haze, drying and conditioning before processing were not significant either. In the case of PBAT-based blends, the LDPE contaminant had no effect on haze. However, the haze of the LDPE-based blends increased with increasing weight fractions of PBAT. Also, LDPE and PBAT are semi-crystalline polymers, therefore an increasing amount of dispersed PBAT particles in the LDPE matrix causes increasing refraction inside the sample, which increases the haze of the blends compared to pure LDPE. The increased haze of recycled LDPE can be a problem in packaging applications.

### Compostability of the blends

Table 1 shows the initial and residual mass of the samples, the degree of disintegration, and the decomposition time for LDPE/PBAT blends with different weight ratios. As expected, neither the dried nor the conditioned LDPE-based samples decomposed. No fragmentation was observed for LDPE-based samples, even at 10 wt% PBAT. In contrast, PBAT-based samples were completely disintegrated in 42 days, regardless of the weight fraction of LDPE. There was no difference in the

decomposition time between the dried and conditioned PBAT samples, either. The LDPE in PBAT-based blends was obviously not biodegraded but can be considered biodegraded by the standard as they passed through a 2 mm sieve.

*Table 1 Initial and residual mass, the degree of disintegration, and the decomposition time for LDPE/PBAT blends with different weight ratios*

	The weight fraction of PBAT/LDPE [wt/wt%]	Initial mass before composting [mg]	Residual mass after composting [mg]	Degree of disintegration [%]	Decomposition time [day]
Dried	100/0	20	0	100	42
	99/1	17	0	100	42
	97.5/2.5	21	0	100	42
	95/5	16	0	100	42
	90/10	14	0	100	42
	0/100	12	12	0	-
Conditioned	100/0	17	0	100	42

The biodegradation process is shown in *Table 2* for LDPE-contaminated PBAT-based blends. We did not observe visible fragmentation and impairment of the samples during the first 24 days. With the exception of pure LDPE, the samples were broken up into small pieces by 35 days and completely degraded by day 42.

*Table 2 Biodegradation steps during composting for PBAT-based blends and pure LDPE*

	The weight fraction of PBAT/LDPE [wt/wt%]			
	100/0	97.5/2.5	90/10	0/100
Day 0				
Day 3				
Day 25				
Day 32				
Day 37				
Day 42				
Scale bar				

## Summary

We analyzed the tensile and optical properties and the compostability of different PBAT/LDPE blends. We found that 1 wt% "contaminant" in LDPE and 2.5 wt% "contaminant" in PBAT do not affect tensile strength and elongation at break. However, 10 wt% impurity decreased elongation at break from 188% to 79% in the LDPE-based and from 307% to 109% in the PBAT-based blends. The LDPE "contaminants" did not affect the optical properties of the PBAT, but the PBAT impurities increased haze from 13% to 23% in the LDPE blends, which can be a problem in packaging applications. We did not observe visible fragmentation and impairment of the films during the first 24 days during composting. The PBAT-based samples were broken up into small pieces by day 35 and completely degraded by day 42.

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