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The effect of biodegradable polymer blending on the disintegration rate of PHBV, PBS and PLA in soil

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ABSTRACT

This study generates new insights into the disintegration phenomena that take place upon blending different classes of biodegradable polymers. Polymer blending is found to be an effective method to tailor the disintegration rate of these polymers in soil. It is shown that the biodegradation of poly(hydroxybutyrate-co-hydroxyvalerate) (PHBV) can be accelerated by blending with poly(butylene succinate-co-adipate) (PBSA) and polycaprolactone (PCL). The observed high rate of disintegration of poly(butylene succinate) (PBS) in soil (severe deterioration in 4 weeks, and fragmentation in 4 months) does not fully align with its current reputation in the market as a polymer that is non-biodegradable in soil. Disintegration trials executed in soil media with different inoculants demonstrate that the biodegradation rate of PBS in soil is highly dependent on the specific soil conditions. Moreover, it is shown that the biodegradation of PBS can be substantially accelerated by blending it with PBSA (fragmentation in 8 weeks). Finally, it is shown that the disintegration of poly(lactic acid) (PLA) in soil can be enhanced by blending it with PCL. Experiments that monitor the CO_2 evolution of these blends, both in soil and in home composting environments, demonstrate that not just the disintegration, but also the overall biodegradation of PLA is enhanced by blending with PCL (39 % conversion to CO_2 in 12 months incubation in soil; 89 % conversion to CO_2 in 6 months incubation in home composting conditions). This opens up possibilities for targeted blending strategies to reduce potential accumulation of PLA-based plastics in soil environments.

1. Introduction

Over the past decades the production and use of plastic materials and products has increased tremendously, thereby enabling the development of more efficient and lightweight applications in transport, packaging, apparel, agriculture and many other domains [1–4]. One of the main assets of plastic materials is that their mechanical, thermal and optical properties can be enhanced by using functional additives or by blending different thermoplastic polymers [5–9]. Another outstanding characteristic of plastic materials is their mechanical, chemical and thermal stability under a wide range of environmental conditions. Unfortunately these properties that make plastics superior materials for many products during use, are also the reason for the largest challenges that plastics face today during their end-of-life. The use of additives and blending of different polymers is amongst the most prominent causes that plastics are more difficult to effectively recycle than other materials such as paper and glass [4,10–14]. Furthermore, their high stability makes that plastic products that end up in nature, will accumulate in the form of plastic product debris and smaller fragments that can no longer be observed by the human eye, so called micro- and nanoplastics [15,16]. The use of biodegradable polymers in applications where leakage into nature is unavoidable (due to abrasion or intended use in soil) can help to reduce the accumulation of plastic materials in the environment [17,18]. Examples of such applications are garments and agricultural cultivation aids [12].

The biodegradation of plastics does not only depend on the chemistry of the polymer, but also on the presence and activity of the biological systems involved in the process. When investigating the biodegradability of a material, the effect of the environment cannot be neglected. For example, poly(lactic acid) (PLA) is a polymer with high biodegradation rate in an industrial composting environment while at the same time its biodegradation in soil is known to be relatively slow [19,20]. Numerous ways for the experimental assessment of polymer biodegradability have been described in the scientific literature [21]. Because of slightly different definitions or interpretations of the term ‘biodegradability’, the different approaches are not always equivalent in terms of information they provide or with respect to their practical significance. Monitoring (visual) disintegration or weight loss is often used as a screening tool to assess the biodegradation of plastics in its field of application. However, it cannot be used to prove its complete microbial metabolic utilization. In case of blended compounds or copolymers, complete disintegration can be achieved by the biodegradation of one of

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the components, also when other constituents do not biodegrade. For demonstrating complete biodegradation of the compound on a chemical level, additional assessment of conversion of polymeric carbon to carbon dioxide is required.

In order to minimize the environmental footprint of biodegradable plastics, it is of high importance that the biodegradation profile of these polymers aligns with both the functionality that is required during use, and the environment in which it is most likely to accumulate [22]. For example, a biodegradable fertilizer coating will require a relatively slow and linear biodegradation profile while garments will need to maintain their full integrity during use and biodegrade as soon as possible upon accidently entering the biosphere. This implies that different biodegradable polymers need to be selected for different applications and that it should be possible to program the biodegradation rate for a specific application-environment combination. In recent years multiple strategies to modify the biodegradation rate of polymers have been proposed of which co-polymerization, use of specific additives and natural fillers, and polymer blending are the most prominent [8,23–27]. Among these approaches polymer blending is most interesting from a cost and environmental point of view. Co-polymerization requires substantial investment in new polymer development and the use of additive route could lead to the presence of other unwanted entities in soil or sea.

To contribute to the collective knowledge on the biodegradation behaviour of biodegradable polymers and their blends, this study systematically investigates the effect of different polymer blend ratios on their disintegration rate in soil. The main polymers studied are poly (hydroxybutyrate-co-hydroxyvalerate) (PHBV), poly(butylene succinate) (PBS) and poly(lactic acid) (PLA). Within this scope PHBV is the only polymer that is certified as soil biodegradable (i.e. biodegrades completely within a 2 year timeframe). Nevertheless it is of interest to direct the biodegradation profile towards the requirements of specific applications by adding other soil biodegradable components [28,29]. PBS is a polymer that is reported to biodegrade relatively quickly in soil [30], but does not pass the existing certification standards for soil [31]. Finally, PLA is studied as it is known to be only biodegradable under industrial composting conditions and does not biodegrade in soil within a reasonable timeframe. For both PBS and PLA, polymer blending could yield new materials that show improved biodegradation behaviour and could subsequently be certified and contribute to the prevention of plastic accumulation in soil environments. A clear disadvantage of formulating new polymer blends for optimized biodegradation profiles is that final level of conversion and subsequent material accumulation is governed by the component with the slowest rate. As an example, blending poly(lactic acid) with soil-biodegradable polymers has been reported to enhance the initial biodegradation onset of the material but typically does not catalyse the biodegradation of the PLA phase [32]. An exemption to this theory was first reported by Narancic et al. [33] who found that a blend consisting of polycaprolactone (PCL) and PLA can meet the requirements for home-composting while other polymers such as semi-crystalline polyhydroxyalkanoates (PHAs) do not yield a similar effect. Another biodegradable polymer that has been reported to accelerate the biodegradation of other polymers upon blending is poly (butylene succinate-co-adipate) (PBSA). Most investigations focussed on combining this polymer with PBS which is a logical result from the molecular similarities between these two polymers which results in materials that display a well-balanced set of properties that relates to the blend ratio selected [24,34]. This study generates new insights on the disintegration phenomena that take place upon blending PHBV, PBS and PLA with PCL and PBSA. The results can therefore serve as a platform for further biodegradable polymer blend development and plastic products that require a specific programmed biodegradation rate.

2. Experimental

2.1. Materials

Poly(butylene succinate) (PBS) BioPBS FZ91PB (M_w = 165 kg/mol) and poly(butylene succinate-co-adipate) (PBSA) BioPBS FD92PB (M_w = 190 kg/mol) were obtained from PTT MCC Biochem. Poly(caprolactone) (PCL) Capa™ 6500D (M_w = 93 kg/mol) was obtained from Ingevity. Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) ENMAT Y1000 (M_w = 305 kg/mol) was obtained from PHARADOX by Helian Polymers. Two grades of poly(lactic acid) being Luminy® LX175 (M_w = 126 kg/mol) (PLA type 1) and Ingeo® 4043D (M_w = 178 kg/mol) (PLA type 2) were obtained from TotalEnergies Corbion and NatureWorks respectively.

2.2. Material preparation

All polymers were dried for at least 8 h in a Gerco two-chamber dry-air desiccant dryer (TTM 2/100 ES, Gerco Kunstofftechnik GmbH, Warendorf, Germany) to achieve moisture contents below 250 ppm prior further processing. The drying temperature was 83 °C, except for PCL which was dried at 40 °C. Polymer blends were prepared via extrusion using a Berstorff ZE25x40D twin screw co-rotating extruder at maximum temperatures of 175 °C, 170 °C, 195 °C for the PHBV, PBS and PLA blends respectively. Test samples for disintegration tests were prepared via sheet extrusion using a Dr. Collin Teachline E20T single screw extruder with a sheet die at maximum temperatures of 175 °C, 180 °C, 195 °C for the PHBV, PBS and PLA blends respectively.

2.3. Biodegradation characterization

Assessment of the disintegration of materials in soil was based on the method described in ASTM G160-12(2019) [35]. Dumbbell shaped samples were cut from sheets (thickness 200 μm) and were exposed to soil under laboratory controlled conditions. The soil, a sandy topsoil obtained from organic experimental and training Farm 'Droevendaal' (Part of Wageningen University & Research, NL), was inoculated with 2 % (based on dry weight) fresh, mature compost from an industrial composting facility treating the organic fraction of municipal solid waste. Containers (40 x 16 × 18 cm) with buried samples were incubated at constant temperature (25 ± 2 °C) and relative humidity (90 ± 5 %). Moisture content of the soil was maintained at 80 % of the water holding capacity by spraying demineralized water to correct for any evaporation during the incubation. Viability of the soil was regularly checked with an untreated cotton cloth reference which should lose >50 % of the initial tensile strength in 5 days of exposure to the soil. In addition, soil pH was monitored as well. Plastic samples were recovered from the soil at regular intervals, for this purpose 6 replicate samples were buried per datapoint. Recovered samples were carefully rinsed and cleaned to remove the soil and subsequently photographed. Prior to mechanical analyses the samples were conditioned for 1 week at 50 % RH and 20 °C. Control samples were recovered after 1 day of exposure to exclude any influence of the burial and cleaning procedure. In addition, specific samples were subjected to gravimetric measurements of carbon dioxide to assess the biodegradation/mineralization, both in home composting conditions (according to ISO 14855 at 28 °C) [36] and in soil conditions (according to ISO 17566 at 25 °C) [37]. The percentage of biodegradation is calculated as the percentage of solid carbon of the test substance that had been converted to mineralized carbon in the form of CO_2. For testing in home composting conditions, the evolved CO_2 was measured directly in the exhaust air of the reactor with a gas chromatograph (PerkinElmer Clarus 500), while the flow rate is measured with a Brooks 5860S mass flow meter. After 47 days the test was converted from dynamic (active aeration) to static (passive aeration) composting conditions. For testing in soil conditions, the evolved CO_2 was captured in KOH and the CO_2 production was regularly determined by

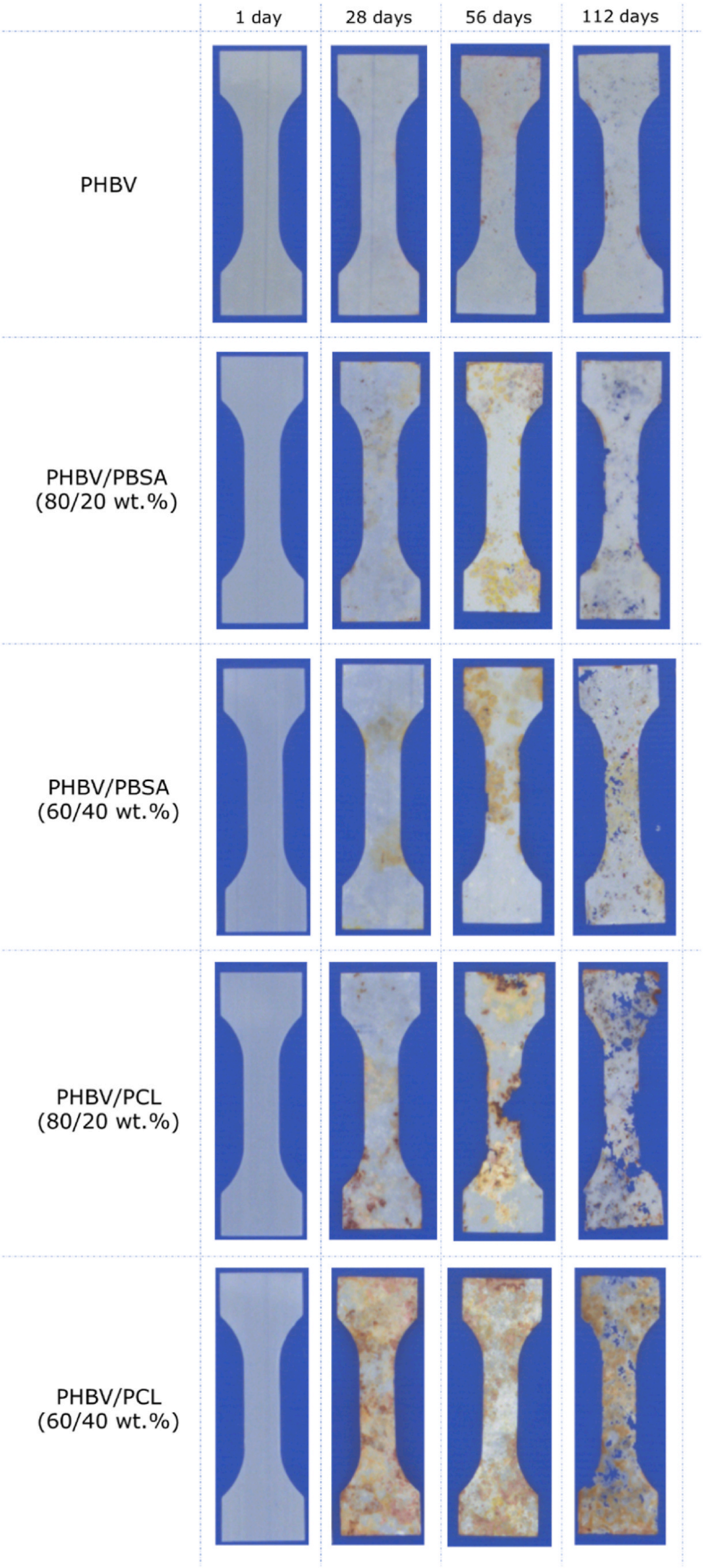

Fig. 1. Examples of specimens (sheet thickness 200 μm) of extruded PHBV blends recovered after 1–112 days of exposure to soil.

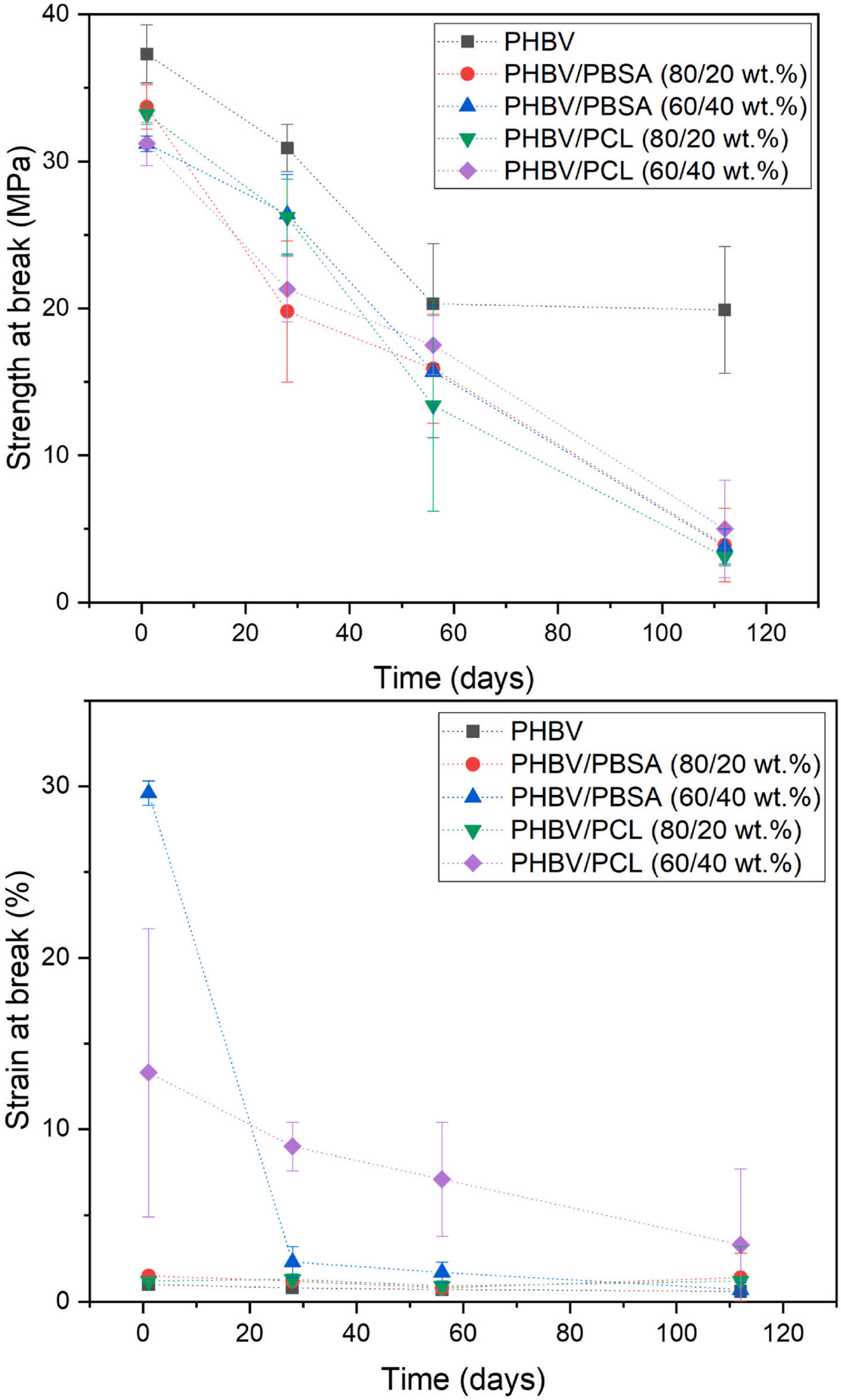

Fig. 2. Mechanical properties of extruded PHBV blends recovered after 1–112 days of exposure to soil.

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2.4. Polymer characterization

Mechanical properties of the polymer blends before and after exposure to soil were evaluated using a Zwick Z010 tester with Multisens extensometers according to ISO 527 method 3 [38]. Measurements were performed in 5-fold.

The molecular weight of polymer samples was determined with gel permeation chromatography (GPC) using a Viscotech VE 2001 GPC max provided with a TDA305 Triple Detector Array (RALLS + LALLS, RI Detector and Viscometer). Columns used were a PSS PFG analytical linear M and guard column, molecular range ~250–2.5·10^6 Da (PMMA in HFIP). The selected solvent was hexafluoroisopropanol (HFIP) with 0.02 M potassium trifluoroacetate (KTFA). All measurements were performed in duplicate.

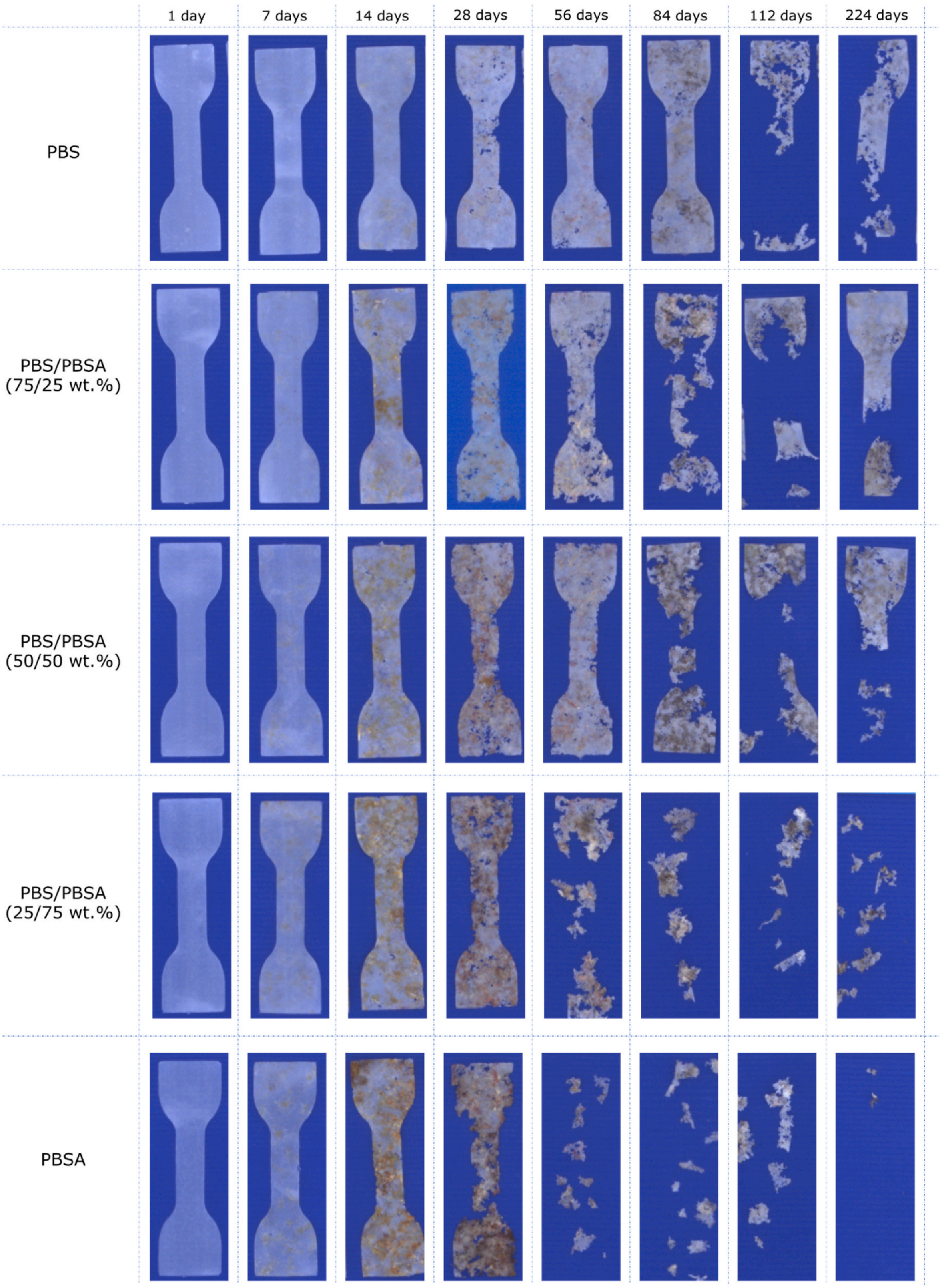

Fig. 3. Examples of specimens (sheet thickness 200 μm) of extruded of PBS/PBSA blends recovered after 1–224 days of exposure to soil.

Differential scanning calorimetry (DSC) measurements were performed using a PerkinElmer DSC 8000 provided with liquid nitrogen cooling and an autosampler. Stainless steel DSC cups with rubber rings were used. Samples were heated from −60 °C to 200 °C with a heating rate of 10 °C/min.

3. Results & discussion

3.1. Biodegradation of PHBV blends

The biodegradation of PHBV-based compounds was assessed by

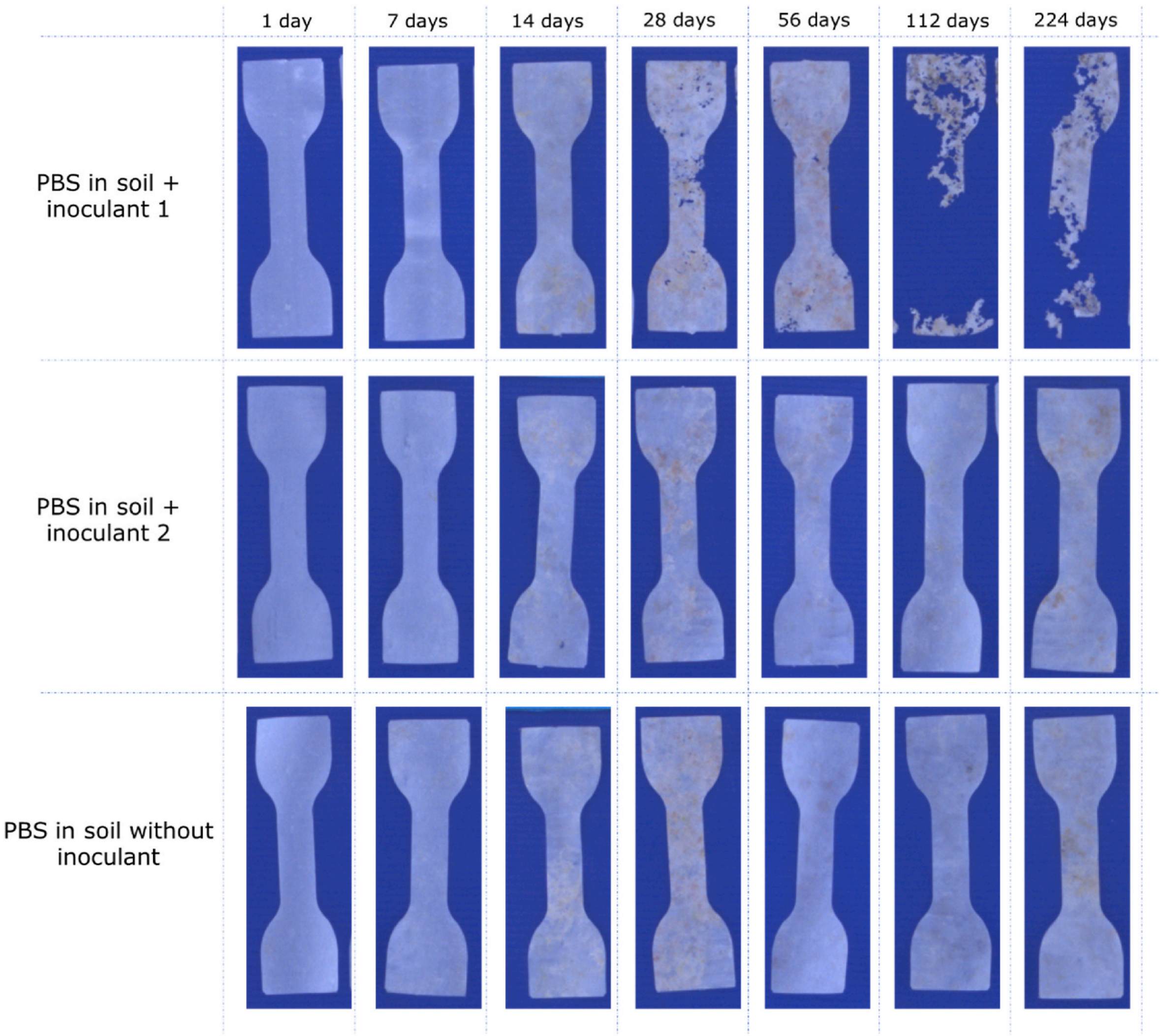

Fig. 4. PBS exposed to soil with different inoculants. Inoculant 1: 2 % mature (but biologically active) compost obtained from an industrial facility processing the organic fraction of municipal solid waste; Inoculant 2: 2 % compost obtained from an outdoor static windrow processing greenhouse waste (dormant due to winter conditions).

exposing them to soil under laboratory controlled conditions and following their disintegration in time. Fig. 1 shows examples of samples that were recovered after up to 112 days of exposure. PHBV samples maintained their shape and integrity in this period, but showed substantial signs of biodeterioration. The samples exposed for 28 days had coloured spots on the surface and deeper into the material, which was more prominent in samples with longer exposure times. In addition, several holes were observed in samples exposed for 58 days, and even more so in samples recovered after 112 days of exposure. This was in line with the gradual decrease in mechanical properties (e.g. strength at break and strain at break) observed with increasing soil exposure time, as shown in Fig. 2. In this respect it should be noted that the experimental data from the mechanical tests show some deviations between replicates, in particular visible for samples with high elongation properties. Given that degradation will cause non-uniform defects along the samples, that is likely to additionally contribute to data scattering.

Compounds of PHBV blended with PBSA showed more pronounced biodeterioration upon exposure to soil (Figs. 1 and 2). With increasing PBSA content, the discoloration after 28 days was more severe, and also the size and number of holes increased in the samples recovered after 58 or 112 days. Discoloration, formation of holes and fragmentation of samples was even more clearly observed for compounds of PHBV blended with PCL. These visual observations indicate that increasing the content of PBSA in PHBV compounds accelerates the material disintegration in soil resulting from polymer biodegradation. When PHBV is compounded with PCL instead of PBSA, the disintegration rate is increased even more.

These results show that it is possible to tailor the disintegration rate in soil by altering the polymer composition of the material. This feature can prove to be advantageous in, for example, greenhouse cultivation processes that require a plastic support in a soil environment for a specific time interval. If the time interval depends on the crop that is grown or seasonal effects, a slight alteration of the material composition can aid the end-of-life scenarios of these products without hampering their functionality.

3.2. Biodegradation of PBS-PBSA blends

In a similar set-up we investigated the biodegradation of PBS-PBSA based materials in soil by following their disintegration in time. Examples of recovered samples are shown in Figs. 3 and 4. In the case of unmodified PBS, some discoloration was already observed in samples recovered after 14 days of exposure to soil and after 28 days, some holes were formed. After 112 days of exposure, all PBS samples had physically disintegrated and only fragments could be recovered. The disintegration of these PBS samples progressed markedly faster than that of the PHBV

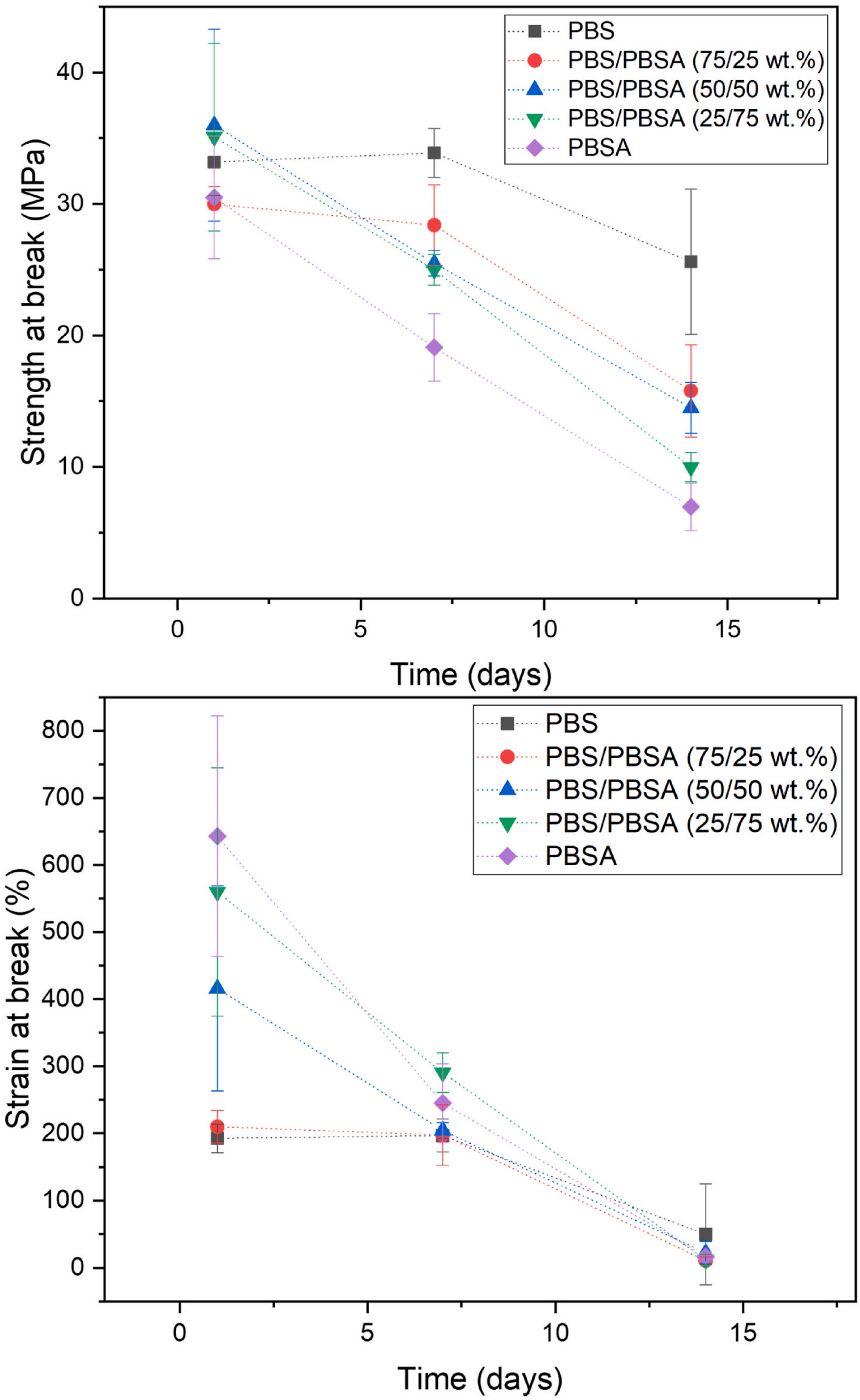

Fig. 5. Mechanical properties of PBS/PBSA blends recovered after exposure to soil.

samples in comparable conditions. This is remarkable because the selected PHBV material is generally recognized as ‘soil biodegradable’ [31] and certified according to ‘OK compost HOME’ and ‘OK biodegradable MARINE’ [39] whereas the particular PBS grade is not.

In the same soil burial test, samples of PBSA disintegrated significantly faster than the corresponding PBS samples; after 14 days of exposure substantial deterioration was observed and after 28 days all 6 replicate samples were severely damaged and of roughly half the specimens only partial fragments could be recovered. These results are in accordance with that PBSA, being chemically similar to PBS but in which part of the succinate moieties is replaced by adipate, is generally recognized as biodegradable in soil conditions [31] and is certified ‘OK biodegradable SOIL’ [40].

Next to PBS and PBSA, Fig. 3 also shows recovered samples of compounded blends of PBS and PBSA with different PBS/PBSA ratios. The rate at which deterioration and fragmentation (disintegration) occurred increased with increasing PBSA content. This is not only reflected in the time it took for samples to disintegrate in soil, but was also

Table 1
Molecular weight of extruded PBS/PBSA blends recovered after 1–112 days of exposure to soil.

Sample	Molecular weight - M_w (kg/mol)						
	Time of exposure						
	1 day	7 days	14 days	28 days	56 days	84 days	112 days
PBS	156	152	148	147	143	132	122
PBS/PBSA (75/25 wt%)	157	154	148	145	137	129	122
PBS/PBSA (50/50 wt%)	159	156	148	147	138	130	118
PBS/PBSA (25/75 wt%)	157	156	146	149	140	132	122
PBSA	201	198	179	171	145	137	135
Abiotic controls (stored at 20 °C and 30 % RH)							
PBS	156	159		154			133
PBSA	201	185		169			144

Table 2
Thermal properties of extruded PBS/PBSA blends before and after 56 days of exposure to soil.

Sample	Before exposure				After 56 days of exposure to soil			
	Crystallization		Melt peak		Crystallization		Melt peak	
	ΔH (J/g)	T (°C)	ΔH (J/g)	T (°C)	ΔH (J/g)	T (°C)	ΔH (J/g)	T (°C)
PBS	−8.7	89.8	67.9	113.0	−8.6	89.8	69.3	113.1
PBS/PBSA (75/25 wt%)	−7.1	84.9	61.1	113.2	−7.3	82.2	61.5	113.9
PBS/PBSA (50/50 wt%)	−4.5	80.2	51.1	112.3	−5.0	78.7	52.5	114.8
PBS/PBSA (25/75 wt%)	−2.1	72.4	40.6	108.1	−3.2	72.3	41.2	115.1
PBSA	−1.2	64.2	36.9	83.2	−1.1	67.0	32.8	85.8

observed before actual fragmentation in the decrease of tensile strength or strain at break (Fig. 5), although substantial deviations in the experimental mechanical data of the recovered samples makes these observations less distinct. Nevertheless, the results imply that the disintegration rate of the extruded product can be tailored to the application needs by adjusting the PBS/PBSA ratio.

In addition to visual inspection and mechanical testing, the molecular weight and thermal properties of the samples were determined after their respective exposure time in soil. Results of these analyses are depicted in respectively Tables 1 and 2. It is found that the molecular weight (M_w) of the PBS-based test specimens decreased with increasing soil exposure time from 156 kg/mol to 122 kg/mol after 112 days in the soil. The M_w of PBSA test specimens measured 201 kg/mol before exposure and 135 kg/mol after 112 days in the soil. Comparison with the visual disintegration that is observed in Fig. 3 shows that even fragments of severely disintegrated test specimens consisted of reasonably high molecular weight polymer. The relatively low decrease in molecular weight during the soil incubations does seemingly not correspond to the molecular weights of materials that are in the end of their disintegration process (<5 kg/mol). This can be explained by the fact that these analyses can only be performed on those parts of the sample that did not undergo extensive biodegradation and disintegration yet. After all, material depolymerized to the level that results in loss of physical integrity and fragmentation is not retrievable from the soil in particle form. This is supported by the observation that the decrease of M_w over time seen for PBS-PBSA blends exposed to soil was in the same range as the decrease in M_w of abiotic control samples stored in a climate chamber (in the dark at 20 °C and 30 % relative humidity) for the same period of time (Table 1). All abiotic control specimens were still completely intact and showed no visual signs of disintegration. Consequently the molecular weight decrease observed over time for all PBS based specimens in this study is attributed to general polymer aging. The limited differences in thermal properties as determined with DSC between starting materials and tiny fragments of largely disintegrated test specimens after soil exposure supports these conclusions. Overall, these tests show that the characterization on a molecular level of recovered particles is a less suitable parameter to follow the biodegradation process of polymers.

Nevertheless, considering the observed deterioration of PBS during soil exposure, its substantial fragmentation in approximately 4 months, supplemented with optical microscopy images of the deteriorated samples (Fig. 6) we conclude that active biodegradation of PBS occurs in soil. This is in line with other reports in literature on the biodegradability of PBS (e.g. Nelson et al. [30], and Šerá et al. [41]). However, there are also studies that conclude that PBS is not sufficiently biodegradable in soil (e.g. Narancic et al. [33]). We can explain this controversy by recognising that the biodegradation rate of PBS is particularly sensitive for the choice of soil inoculum. This is illustrated in Fig. 4, showing recovered PBS samples from 3 different experiments in which the soil matrix was supplemented with compost of different origin. When the used standard soil was inoculated with 2 wt% of mature (but biologically active) compost obtained from an industrial facility processing the organic fraction of municipal solid waste, disintegration of PBS occurred much faster than when the compost inoculum was omitted. Supplementing the soil with 2 % of inoculum obtained from an outdoor static windrow processing greenhouse waste (dormant due to winter conditions) also resulted in a lower disintegration rate of PBS. It should be noted that the sensitivity of the disintegration rate for the soil inoculum was not observed for PHBV (and PCL) based compounds (pictures not shown). These observations are in line with a recent study by Lee et al. [42] in which the biodegradation rate of biodegradable polymers was assessed in two different soils by measuring weight loss of film samples. They reported negligible weight loss in 6 months for PBS film buried in a standard horticulture topsoil, whereas in the same period weight loss was complete for the same PBS film buried in a fertilized topsoil (inoculated with vermi-compost). It will be interesting

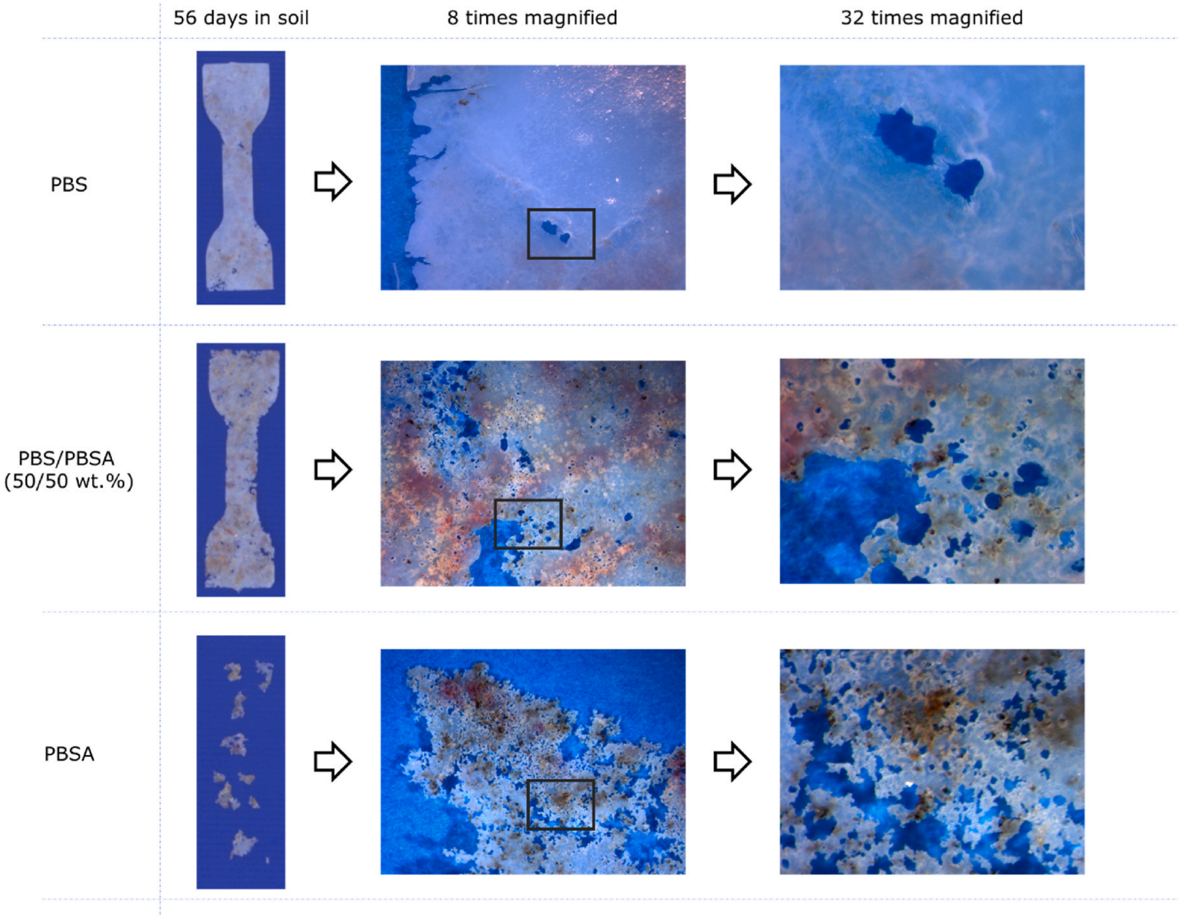

Fig. 6. Optical microscopy evaluation of 200 μm thick specimens of PBS/PBSA compounds recovered after 56 days of exposure to soil. Magnification: 8x and 32x of the same sample.

to explore whether the selection of soil inoculum source similarly affects the biodegradation rate of PBS in the standard respirometric method for the assessment of polymer biodegradability in soil (ISO 17556 [37]). This is the test method currently used to demonstrate compliance with the requirements for a material to be certified 'biodegradable in soil' [40,43].

3.3. Biodegradation of PLA-based blends

A third group of materials investigated for their biodegradability in soil comprises blends of PLA with the soil degradable polyesters PBSA and PCL. Figs. 7 and 8 show representative specimens of PLA-based compounds recovered after exposure to soil for up to 11 months. The pure PLA reference samples did not show visible signs of deterioration after 336 days of soil exposure. This was the case for both grades of PLA, and furthermore, no significant changes in tensile properties were observed in this period (Fig. 9). This was expected because, although PLA is rapidly biodegradable under industrial composting conditions, it is known to degrade only very slowly under ambient temperature conditions [19].

The PLA compounds containing 20 wt% PBSA also did not show deterioration during 336 days of soil exposure. The blends appeared less transparent than the pure PLA materials, probably due to imiscibility of the polymers resulting in some phase separation. Although no visible deterioration was observed for the PLA/PBSA 50/50 blend, it lost its initial high flexibility (strain at break = 215 ± 123 %) in the first 25 days of incubation in soil. Also its tensile strength decreased gradually over time, in contrast with the PLA containing just 20 wt% or 0 wt% PBSA (Fig. 9). Upon comparison of Figs. 7 and 8, no differences in the disintegration behaviour of PLA types 1 and 2 and their corresponding blends were observed.

The PLA/PCL 50/50 blend, however, demonstrated significant different disintegration behaviour. Samples showed substantial discoloration already after 25 days of exposure to soil and at the next sampling moment (at 84 days) all 6 replicate specimens were disintegrated and only fragments could be recovered. Because this was not observed for the PLA/PCL 80/20 blends, we speculate that in this case the PCL fraction is embedded in a continuous phase of PLA which prevents easy access of PCL for the enzymes or soil microorganisms. It has to be noted that these results do not confirm the actual biodegradation of the blend as a whole in soil conditions. It could very well be that only the PCL polymer biodegrades and the PLA persists in soil in particulate form that can no longer effectively be recovered from the soil.

Our observations on the disintegration of PLA/PCL 80/20 blends in soil are in line with the work of Narancic et al. who reported a very low level of biodegradation for a comparable blend of PLA and PCL [33] following the standard test method for assessing biodegradation in soil [37]. Based on their test result, they estimate that this particular PLA/PCL blend wil have a residence time of roughly 33 years in an unmanaged soil environment. Remarkably, they reported in contrast a very high level of biodegradation of the same PLA/PCL blend when assessed in home composting conditions, from which was concluded that a PLA/PCL 80/20 blend can be classified as home compostable, but not soil degradable.

To obtain a better understanding of the biodegradation behaviour of the PLA/PCL 80/20 blend, gravimetric measurements of carbon dioxide were performed, both in home composting conditions (according to ISO 14855 at 28 °C) [36] and in soil conditions (according to ISO 17566 at

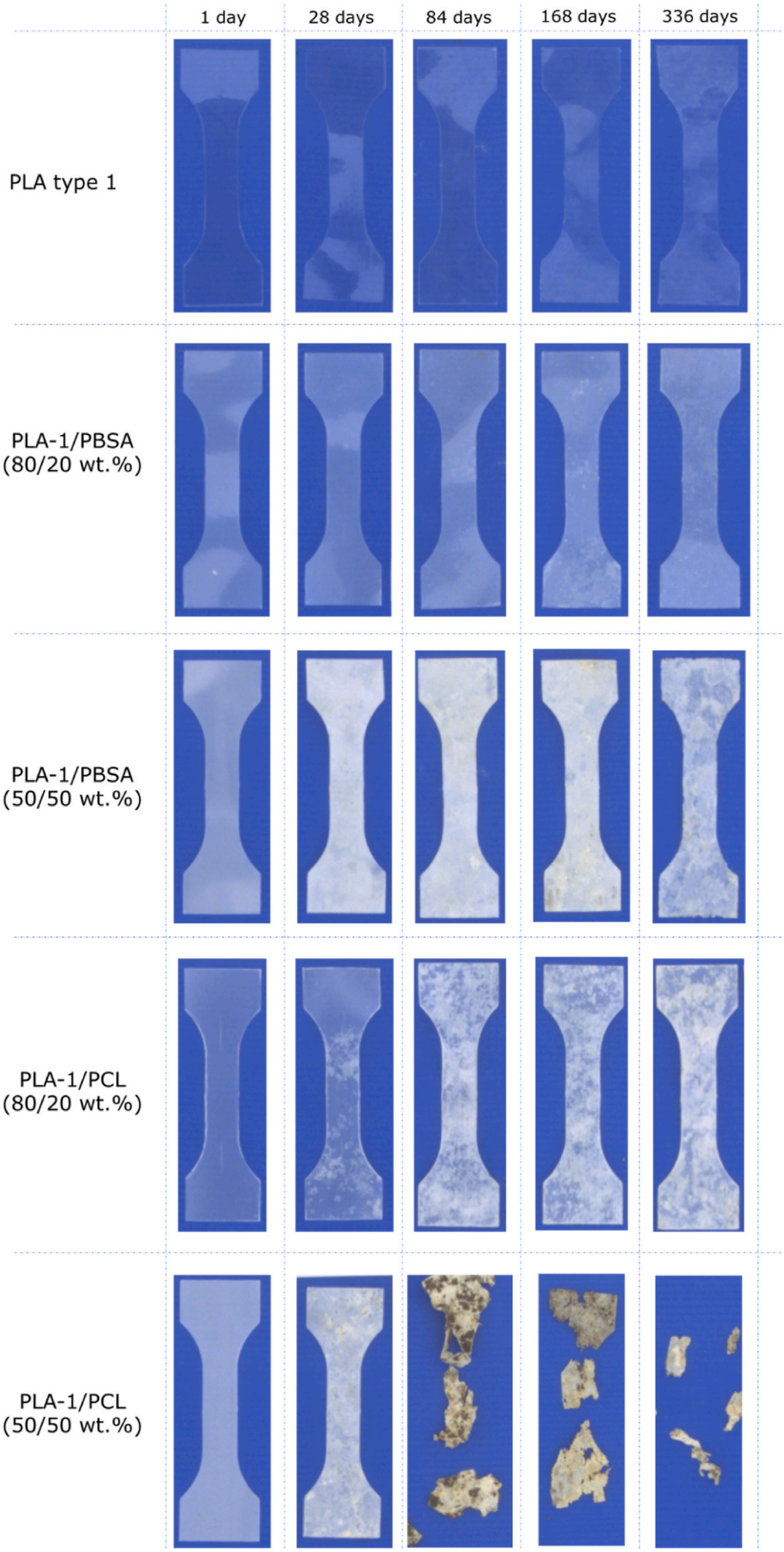

Fig. 7. Examples of specimens (sheet thickness 200 μm) of extruded PLA type 1 blends recovered after 1–336 days of exposure to soil.

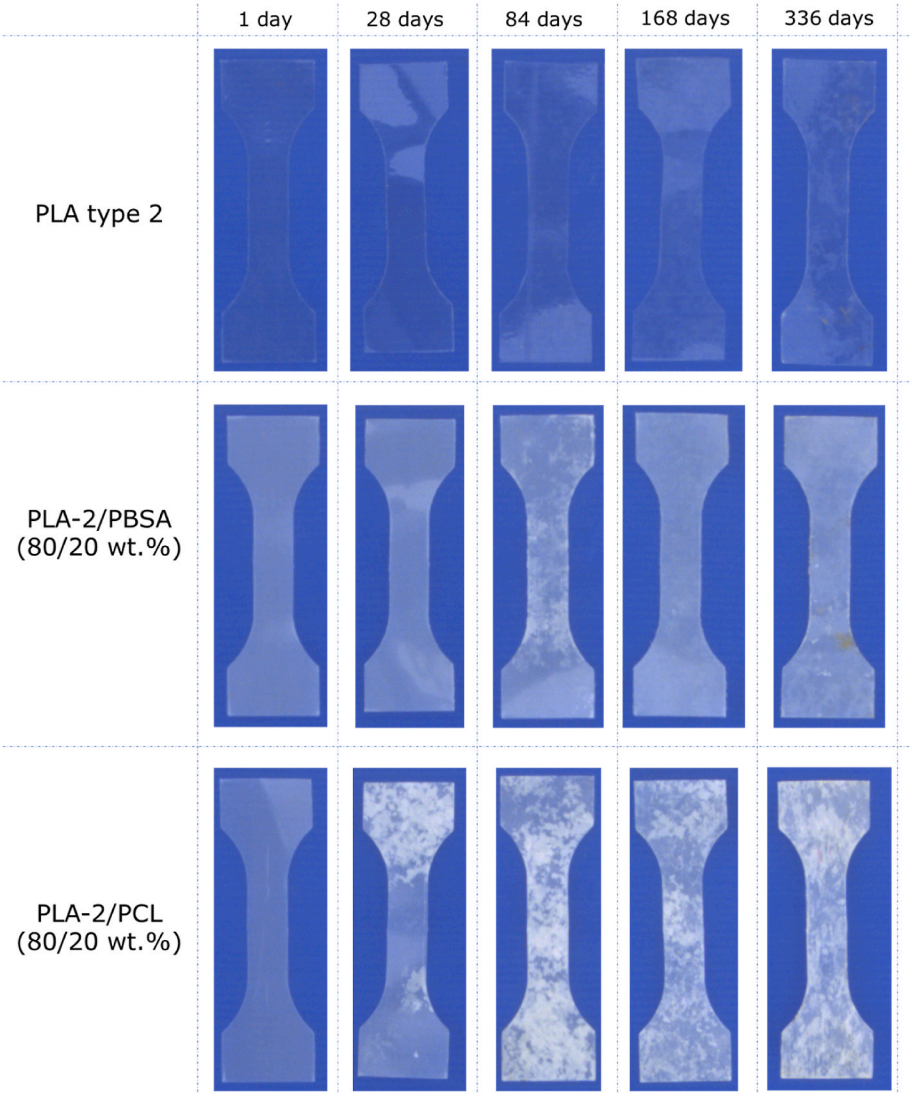

Fig. 8. Examples of specimens (sheet thickness 200 μm) of extruded PLA type 2 blends recovered after 1–336 days of exposure to soil.

25 °C) [37], which are depicted in Fig. 10. As the CO_2 evolution under home composting conditions almost overlaps with that of the reference cellulose material, these results clearly indicate the relatively fast and complete biodegradation of this material composition, thereby affirming the conclusion of Narancic et al. [33] regarding home compostability. However, in Fig. 10, it is also shown that over the timespan of one year the same material reaches a level of almost 40 % biodegradation in soil which is notably higher than Narancic et al. observed in soil. This implies that not only the PCL fraction was mineralized, but also some of the carbon from the PLA polymer was metabolized into CO_2. Based on extrapolation of the observed trend it can be speculated that the extent of biodegradation in 2 years, may possibly even be sufficient to pass the threshold for the 'soil biodegradable' classification.

To explain observed low biodegradation rates of PLA in ambient conditions, it is often argued that the temperature of the degradation environment should exceed the T_g of PLA (~58 °C) to enable hydrolysis of the polymer facilitating subsequent biodegradation [19,44]. Considering that in Fig. 10 the test temperatures are more or less the same and that the main differences between the soil and home composting environments are the inoculum and the (quantity of the) microbial population, there are apparently more factors involved that can enhance the biodegradation of PLA at ambient temperatures. This is affirmed by Han et al. [26] who recently reported that the biodegradation of PLA in home composting conditions was accelerated by blending it with amorphous polyhydroxyalkanoate (PHA). Although more research is required to fully understand the underlying mechanisms, our results indicate that blending PLA with other readily biodegradable polymers can serve as a platform for development of plastic products that require a specific programmed biodegradation rate.

4. Conclusions

Polymer blending is found to be an effective method to tailor the disintegration rate of PBHV, PBS and PLA in soil. It is found that the biodegradation of PHBV can be accelerated by blending with PBSA and PCL. The rate of disintegration of PBS in soil reported in this work is

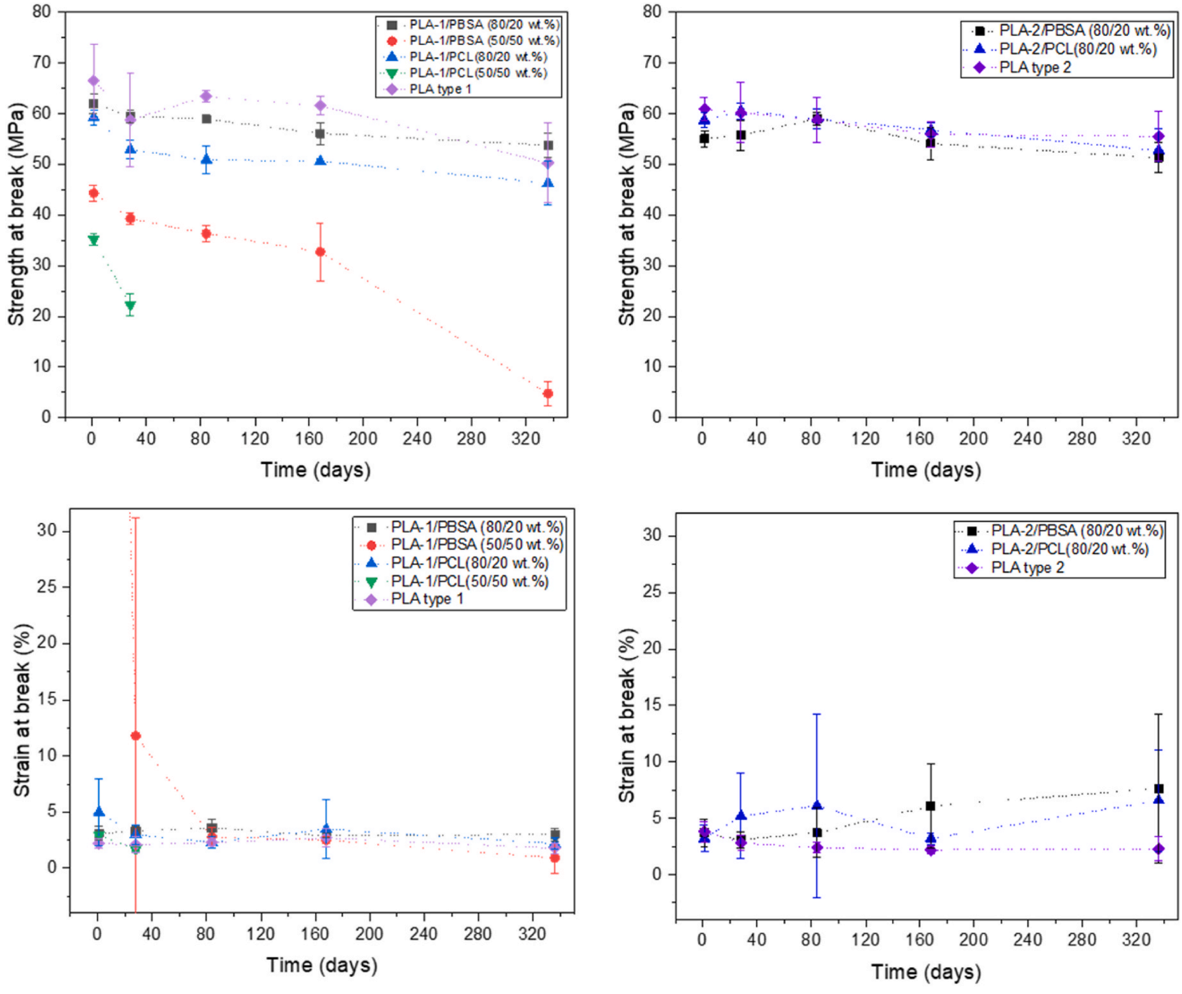

Fig. 9. Mechanical property evaluation of PLA blends after 1–336 days of exposure to soil (left: PLA type-1 blends, right: PLA-type 2 blends).

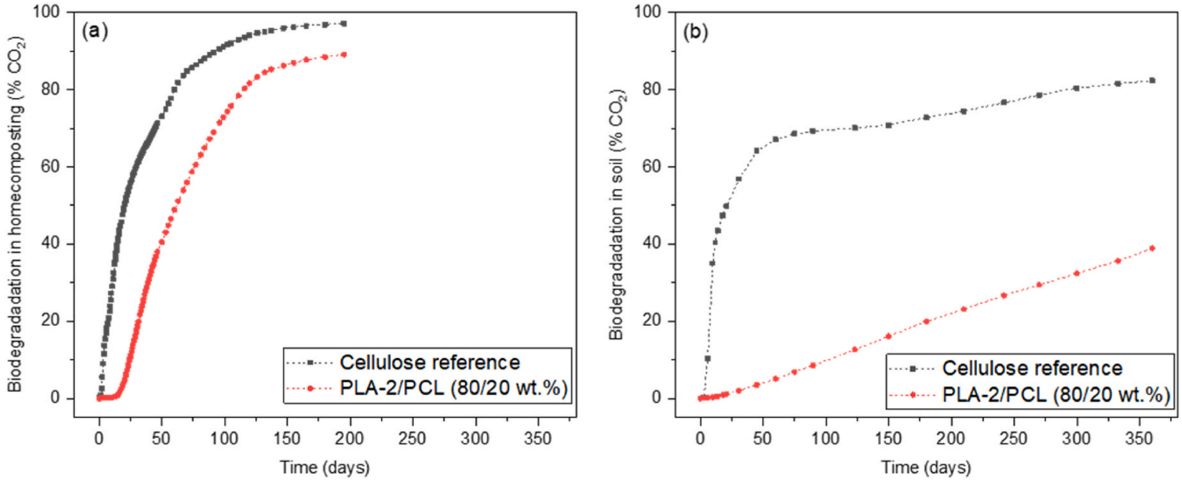

Fig. 10. Biodegradation of PLA type 2/PCL (80/20 wt%) blend under (a) home composting conditions (ISO 14855 at 28 °C) and (b) soil conditions (ISO 17556 at 25 °C).

remarkably high which does not fully align with its current reputation in the market as a polymer non-biodegradable in soil. Disintegration trials executed in soil media with different inoculants demonstrate that the biodegradation rate of PBS in soil is highly dependent on the specific soil conditions. Moreover, it is shown that the disintegration of PBS can be substantially accelerated by blending it with PBSA. Finally, it is shown that the disintegration of PLA in soil can be enhanced by blending with PCL. Experiments that monitor the CO_2 evolution of these blends, both in soil and in home composting environments, demonstrate that not just the disintegration, but also the overall biodegradation of PLA is enhanced by blending it with PCL. This opens up possibilities for targeted blending strategies to reduce potential accumulation PLA-based plastics in soil environments.

CRediT authorship contribution statement

Maarten van der Zee: Writing – original draft, Methodology, Formal analysis, Conceptualization. **Martin Zijlstra:** Writing – review & editing, Investigation, Data curation. **Lambertus J. Kuijpers:** Writing – review & editing, Investigation, Data curation. **Marieke Hilhorst:** Writing – review & editing, Investigation, Data curation. **Karin Molenveld:** Writing – review & editing. **Wouter Post:** Writing – original draft, Supervision, Formal analysis, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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References

[1] N. Kawashima, T. Yagi, K. Kojima, How do bioplastics and fossil-based plastics play in a circular economy? Macromol. Mater. Eng. 304 (9) (2019).
[2] World Economic Forum, E.M.F.a.M.C., The New Plastics Economy — Rethinking the Future of Plastics, 2016.
[3] E.M. Melchor-Martínez, et al., Towards a circular economy of plastics: an evaluation of the systematic transition to a new generation of bioplastics, Polymers 14 (6) (2022) 1203.
[4] Z. Zhu, et al., Packaging design for the circular economy: a systematic review, Sustain. Prod. Consum. 32 (2022) 817–832.
[5] K.E. Fagelman, J.T. Guthrie, Polymer blend formulation and processing, with reference to the nature and the behaviour of pigmented polycarbonate -poly (butylene terephthalate) (PC-PBT) blends, Surf. Coating. Int. B Coating. Trans. 89 (1) (2006) 1–14.
[6] D. Mangaraj, Elastomer blends, Rubber Chem. Technol. 75 (3) (2002) 365–427.
[7] L.M. Robeson, Applications of polymer blends: emphasis on recent advances, Polym. Eng. Sci. 24 (8) (1984) 587–597.
[8] W. Post, et al., Effect of mineral fillers on the mechanical properties of commercially available biodegradable polymers, Polymers 13 (3) (2021) 394.
[9] M. Eesaee, et al., Morphology and crystallization behaviour of polyhydroxyalkanoates-based blends and composites: a review, Biochem. Eng. J. (2022) 187.
[10] M.T. Brouwer, et al., Predictive model for the Dutch post-consumer plastic packaging recycling system and implications for the circular economy, Waste Manag. 71 (2018) 62–85.
[11] W. Sikorska, et al., End-of-Life options for (Bio)degradable polymers in the circular economy, Adv. Polym. Technol. 2021 (2021) 6695140.
[12] E. Maaskant, et al., Strategic selection tool for thermoplastic materials in a renewable circular economy: identifying future circular polymers, Sustain. Prod. Consum. 38 (2023) 174–185.
[13] J.N. Hahladakis, et al., An overview of chemical additives present in plastics: migration, release, fate and environmental impact during their use, disposal and recycling, J. Hazard Mater. 344 (2018) 179–199.
[14] A. Dorigato, Recycling of polymer blends, Adv. Indust. and Eng. Polymer Res. 4 (2) (2021) 53–69.
[15] K. Ghosh, B.H. Jones, Roadmap to biodegradable plastics-current state and research needs, ACS Sustain. Chem. Eng. 9 (18) (2021) 6170–6187.
[16] K. Zhang, et al., Understanding plastic degradation and microplastic formation in the environment: a review, Environ. Pollut. 274 (2021) 116554.
[17] M.S. Kim, et al., A review of biodegradable plastics: chemistry, applications, properties, and future research needs, Chem. Rev. 123 (16) (2023) 9915–9939.
[18] M. Cucina, The lesser of two evils: enhancing biodegradable bioplastics use to fight plastic pollution requires policy makers interventions in Europe, Environ. Impact Assess. Rev. 103 (2023) 107230.
[19] Y. Tokiwa, B.P. Calabia, Biodegradability and biodegradation of poly(lactide), Appl. Microbiol. Biotechnol. 72 (2) (2006) 244–251.
[20] N.A. Rosli, et al., Comprehensive exploration of natural degradation of poly(lactic acid) blends in various degradation media: a review, Int. J. Biol. Macromol. 187 (2021) 732–741.
[21] M. van der Zee, Methods for evaluating the biodegradability of environmentally degradable polymers, Handbook of Biodegradable Polymers, De Gruyter, 2020, pp. 1–21.
[22] I. Paul-Pont, et al., Discussion about suitable applications for biodegradable plastics regarding their sources, uses and end of life, Waste Manag. 157 (2023) 242–248.
[23] L. Liu, et al., Biodegradability of poly(butylene succinate) (PBS) composite reinforced with jute fibre, Polym. Degrad. Stabil. 94 (1) (2009) 90–94.
[24] N. Puekpoonpoal, et al., Morphology development of PLAs with different stereoregularities in ternary blend PBSA/PBS/PLA films, Polymer-Plastics Technol. Mater. 60 (15) (2021) 1672–1685.
[25] G. Engler L, et al., Designing sustainable polymer blends: tailoring mechanical properties and degradation behaviour in PHB/PLA/PCL blends in a seawater environment, Polymers 15 (13) (2023) 2874.
[26] G. Han, et al., Biodegradation behavior of amorphous polyhydroxyalkanoate-incorporated poly(l-lactic acid) under modulated home-composting conditions, Polym. Test. 126 (2023) 108155.
[27] E. Pesaranhajiabbas, M. Misra, A.K. Mohanty, Recent progress on biodegradable polylactic acid based blends and their biocomposites: a comprehensive review, Int. J. Biol. Macromol. 253 (2023) 126231.
[28] M.C. Righetti, et al., Immiscible PHB/PBS and PHB/PBSA blends: morphology, phase composition and modelling of elastic modulus, Polym. Int. 71 (1) (2022) 47–56.
[29] M. Kervran, et al., Thermal degradation of polylactic acid (PLA)/polyhydroxybutyrate (PHB) blends: a systematic review, Polym. Degrad. Stabil. 201 (2022).
[30] T.F. Nelson, et al., Biodegradation of poly(butylene succinate) in soil laboratory incubations assessed by stable carbon isotope labelling, Nat. Commun. 13 (1) (2022) 5691.
[31] V. Bauchmüller, et al., BioSinn: Products for Which Biodegradation Makes Sense, Nova Institute, 2021.
[32] S. Yang, et al., Characterization and biodegradation behavior of bio-based poly (lactic acid) and soy protein blends for sustainable horticultural applications, Green Chem. 17 (1) (2015) 380–393.
[33] T. Narancic, et al., Biodegradable plastic blends create new possibilities for end-of-life management of plastics but they are not a panacae for plastic pollution, Environ Sci Technol 52 (2018) 10441–10452.
[34] M. Puchalski, et al., Molecular and supramolecular changes in polybutylene succinate (PBS) and polybutylene succinate adipate (PBSA) copolymer during degradation in various environmental conditions, Polymers 10 (3) (2018) 251.
[35] ASTM G160-12(2019), Standard Practice for Evaluating Microbial Susceptibility of Nonmetallic Materials by Laboratory Soil Burial, ASTM International, 2019.
[36] ISO 14855-2, in: Determination of the Ultimate Aerobic Biodegradability of Plastic Materials under Controlled Composting Conditions, ISO, 2018.
[37] ISO 17556, in: Plastics - Determination of the Ultimate Aerobic Biodegradability of Plastic Materials in Soil by Measuring the Oxygen Demand in a Respirometer or the Amount of Carbon Dioxide Evolved, ISO, 2019.
[38] ISO 527-3, Plastics - Determination of Tensile Properties - Part 3: Test Conditions for Films and Sheets, ISO, 2018.
[39] TÜV AUSTRIA, Certified products, Available from: https://www.tuv-at.be/okcert/certified-products/, 2024.
[40] T.Ü.V. Austria, OK biodegradable SOIL : initial acceptance tests, in: Program OK 10 - Bio Products – Degradation in Soil, TÜV AUSTRIA BELGIUM, 2012.
[41] J. Šerá, et al., Accelerated biodegradation testing of slowly degradable polyesters in soil, Polym. Degrad. Stabil. 171 (2020) 109031.
[42] J. Lee, et al., Mimicking real-field degradation of biodegradable plastics in soil and marine environments: from product utility to end-of-life analysis, Polym. Test. 131 (2024) 108338.
[43] DIN CERTCO, Certification Scheme “Biodegradable in Soil”, DIN Certco, 2023. Berlin.
[44] S.M. Satti, et al., Biodegradation of poly(lactic acid) in soil microcosms at ambient temperature: evaluation of natural attenuation, bio-augmentation and bio-stimulation, J. Polym. Environ. 26 (9) (2018) 3848–3857.