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Recyclability of PLA and PLA-PBAT Compounds

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Abstract

In this study, mechanical recycling of polylactic acid (PLA) was investigated for up to 10 cycles. As expected, PLAs mechanical properties (tensile strength and modulus, charpy impact strength, heat deflection temperature) decreased after multiple rounds of recycling. Thermal properties and melt flow rate were also measured to examine the effect of reprocessing on the molecular structure. PLA was combined with industrial biopolymer waste (IW) containing polybutylene adipate terephthalate (PBAT) in different ratios to investigate the recyclability of potential biopolymer waste streams. The results showed that the properties (rheological, thermal and structural) of the mixed biopolymer waste stream (containing PLA and PBAT) are highly dependent on the ratios of the different polymers. Mechanical properties changed from brittle to tough when PBAT content was increased. These results can be beneficial for manufacturers and recyclers who are trying to incorporate biopolymers into their processes or who are dealing with different biopolymer waste streams at the same time.

Keywords

polylactic acid, polybutylene adipate terephthalate, recycling, industrial waste

1 Introduction

One of the most well-known environmental issues of our time is the large amount of mismanaged plastic waste. These materials are integral parts of our daily lives, and it is hard to imagine life without them. Despite the numerous advantages they offer, such as affordability, easy processing, and good mechanical properties, they also come with well-known disadvantages. Most plastics are produced from fossil fuels, resulting in significant environmental emissions, and due to the stability of their chemical structure, they do not decompose after use, thus polluting the environment as waste [1]. According to statistical data from the Organisation for Economic Co-operation and Development (OECD) [2] in 2019, the plastics industry was responsible for 3.4% of the world's greenhouse gas emissions, which equates to 1.8 billion tons of CO_2. About 90% of these emissions stem from the production of polymers, primarily derived from petroleum.

According to the Nova-Institute's [3] statistics, in 2021 global plastic production reached 390.7 million tons. Of this, 8.3% consisted of post-consumer recycled plastics (PCR), and 1.5% was attributed to biopolymers. The remaining 90.2% was plastics produced from fossil fuels. When considering Europe, the annual total production was 57.2 million tons, with recycled and bioplastic quantities making up 12.4%.

Due to the significance and widespread attention of the issue, numerous efforts have been made to reduce the environmental impact of plastics including the recycling of plastic products [4–7] and developing biobased and biodegradable polymers also known as biopolymers [8–10]. However, the widespread adoption of bioplastics is currently limited by their high cost and inadequate properties [11]. PLA is a biobased and biodegradable polymer that belongs to the group of polyhydroxyalkanoates (PHA). It is a semi-crystalline polymer with a glass transition temperature of around 58 °C and a melting point between 160–180 °C [12]. Among PHAs, PLA is the easiest to process using traditional plastic processing technologies (extrusion, injection molding) and it is also one of the most used biopolymers, taking up 18% of worldwide biopolymer production in 2022 second to starch-containing compounds, according to the report of the European Bioplastics & Nova-Institute [13].

Polybutylene adipate terephthalate (PBAT) is a fossil-based biodegradable polymer [14]. Contrary to other

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biopolymers, PBAT's mechanical properties are more similar to polyethylene. It is a highly flexible material and therefore promising for various applications as is or compounded with biopolymers to make them less stiff [15].

Mechanical recycling is a commonly used method to recycle industrial waste and its relevance is increasing with the appearance of regulations restricting the use of single-use plastics. Among them is the Directive (EU) 2019/904 of the European Parliament and of the Council of 5 June 2019 on the Reduction of the Impact of Certain Plastic Products on the Environment, which aims to ensure that by 2025, PET bottles contain at least 25% recycled plastic [16].

Several studies have shown that fossil-based polymers like polypropylene (PP), polyethylene (PE), and polyethylene terephthalate (PET) can be mechanically recycled. Their degradation mechanism and heat sensitivity may vary, resulting in different recyclability. Schyns and Shaver [7], examined the recyclability of PP and PE and concluded that with recycling the mechanical properties deteriorate and the processability of the materials also decreases. Processability and viscosity in the melt state are crucial parameters from an industrial point of view, for example in the packaging industry, where thin films are extruded and blown.

While biopolymers can degrade in certain circumstances, composting might not be their best end-of-life option. Cosate de Andrade et al. [17], compared the mechanical and chemical recycling and composting of PLA by Life cycle assessment (LCA) and found that mechanical recycling was the most environmentally friendly option. Following the same path Carrasco et al. [18] investigated the properties of an extrusion grade PLA, and found that in three processing cycles, the melt flow rate (MFR) values grew from 7 g/10 min to 10.7 g/10 min, which can influence the extrusion process greatly. Zenkiewicz et al. [19] characterized the properties of multi-extruded PLA. After 0-10 extrusions, the samples to be examined were produced by injection molding. During mechanical tests, it was observed that the tensile modulus and strain at break did not significantly change even after multiple extrusions. Stress at break decreased from 72 MPa to 68 MPa with the number of extrusions. Similarly, impact resistance decreased from the already low value of 2.5 kJ/m^2 to 2.0 kJ/m^2. The MFR values changed to a greater extent, increasing more than 3 folds during recycling. This suggests that the molecular weight of PLA decreased due to multiple processing cycles. Hopmann et al. [20] simulated the incorporation of intermediate production waste by adding different ratios of recycled materials to the production of samples. They found that the mechanical properties did not change significantly, while molecular weight decreased by 28% when they used 100% recycled material.

Few studies focus on the place of PLA in the waste management system. Environmentally, recycling is advised, but the amount of biopolymers used today is not enough for a justified separate collection. The different biopolymers are usually blendable therefore several studies focused on their compounds [21–24]. If they were to be collected together, it is imperative to analyze the properties of the different materials recycled together. La Mantia et al. [25] examined the recyclability of BASF's Ecovio F23B1 grade, which is a blend of PLA and PBAT with 12 w% calcium carbonate. As the number of cycles increased, an increase in viscosity was observed, which was correlated with the branched molecular structure of PBAT. Among the mechanical properties, an increase in elastic modulus and a decrease in elongation at break were observed with the number of extrusions.

While previous studies cover the recyclability of fossil-based polymers and extrusion-type biopolymers, there is a lack of research that focuses on recycling by injection molding and the investigation of the feasibility of a joint collection and recycling of PLA and PBAT.

This work aims to investigate the properties of injection-molded PLA through 10 cycles of processing. Between processing steps, test specimens were ground mechanically and were reprocessed to simulate recycling as it is done industrially. Additionally, the reusability of a PLA-PBAT compound (referred to as industrial waste (IW)) was examined. Different ratios of recycled PLA and the IW were mixed and processed to investigate the joint recyclability of these biopolymers. Injection molded samples were tested for their mechanical (tensile, bending, and dynamical testing), thermal, rheological, and structural properties.

2 Experiments

2.1 Materials

For the investigation of PLA recyclability, PLA IngeoTM Biopolymer (Grade 3025 D) was obtained from NatureWorks Ltd., USA. The material has a density of 1.24 g/cm^3 and a tensile strength of 62 MPa with 3.5% deformation at break. The melt flow rate (MFR) is 14 g/10 min (210 °C, 2.16 kg). To prepare PLA-PBAT blends from production waste BF8515 from Sirmax Group (BioComp, Italy), a compound consisting of 85 w% PLA and 15 w% PBAT was utilized. This compound has a density of 1.21 g/cm^3, its tensile strength is 26 MPa, and the elongation at break is 150%. The MFR is 5 g/10 min.

2.2 Material preparation and processing

For the production of the samples, an Arburg 570 S hydraulic injection molding machine (Arburg GmbH, Germany) was used. The mold has 6 cavities and it is suitable for producing MSZ EN ISO 527 "Type 1A" test specimens. Before injection molding, all materials were dried in a drying cabinet (Taisite FCO-230L, China) at 80 °C overnight.

The grinding step of recycling was performed using a Wittmann Battenfeld G-Max 9 (Wittmann Battenfeld GmbH, Austria) rotary blade grinder. The grinder's sieve provided a maximum particle diameter of 5 mm, which was used for injection molding of the recycled samples. To simulate the mechanical recycling of PLA on its own, the injection molded samples were granulated and remolded again 9 times, so the last batch of material was processed 10 times altogether. In our experiments, PLA granules were also blended with the production waste of a PLA-PBAT compound (IW), which contained 15 w% PBAT and 85 w% PLA.

2.3 Methods

To evaluate the mechanical properties of the produced materials a wide range of mechanical tests were carried out. According to the MSZ EN ISO 527-1 standard tensile tests were conducted on Instron 8850 and Instron 8874 (Instron, USA) mechanical testing machines equipped with 5 kN cells, with 1 mm/min crosshead speed. Both tensile and bending tests were conducted to measure the tensile and flexural modulus and strength. Ceast Resil Impactor Junior (Instron, USA) equipped with a 2 J hammer was used to determine the Charpy impact strength.

Thermal properties and crystallinity were examined by differential scanning calorimeter DSC 250 (TA Instruments, USA). Test specimens were dried for at least 12 hours before measurement at 40 °C. The specimens were first heated from 20 to 200 °C, then cooled to 20 and reheated to 200 °C at a rate of 10 °C/min. Between the cooling and heating phases, 1 minute tempering times were used. The measurements were carried out under dynamic nitrogen atmosphere. To examine the crystalline structure in more detail we also conducted measurements with 5 °C/min and 20 °C/min heating/cooling rates within the same temperature limits.

The rheological behavior of the materials was characterized by Melt Flow Rate (MFR) measurements using an Instron MF20 (Instron, USA) machine. The tests were carried out according to the MSZ ISO 1133 standard at 210 °C and 2.16 kg load.

Scanning electron microscopy pictures were taken with Tescan Vega SEM device (Tescan Group, Czech Republic) to investigate the compatibility of PLA and PBAT blends.

To determine the heat deflection temperature (HDT) a Ceast HV3 6911.000 (Instron, USA) machine was used. The measurements were made under 0.45 MPa stress and 120 °C/h heating rate.

3 Results and discussion

3.1 Processing of the materials

During the mechanical recycling of PLA in itself, most of the injection molding parameters stayed the same as the number of processing cycles increased, as shown in Table 1. However, due to the degradation of the material some of the parameters needed to be altered to manufacture correct specimens. These were: the volume in the barrel before switching over to holding pressure, cooling time, and decompression. All these were due to the gradual decrease of molten PLA's viscosity. Decompression needed to be raised from 7 cm^3 to 15 cm^3, and cooling time was increased from 45 s to 55 s. It is also important to state, that by the 10th cycle of processing some of the specimens broke just by the force of the ejection.

When processing the PLA-IW compounds, the barrel temperature was slightly lowered to 160-180 °C due to PBAT's heat sensitivity. For the injection molding, different ratios of IW and granulated PLA were used in 90/10, 30/70, 50/50, 70/30, and 10/90 w% ratios. Since the IW consisted of 85 w% PLA and 15 w% PBAT the compounds' exact composition can be calculated. The final PBAT and PLA contents are shown in Table 2. To evaluate the miscibility of PBAT and PLA scanning electron microscopy (SEM) was used. In the pictures of compounds containing 10 w% and 50 w% IW (Fig. 1), small fractions of separated material can be seen, which indicates that the two polymers are not miscible.

3.2 Thermal properties

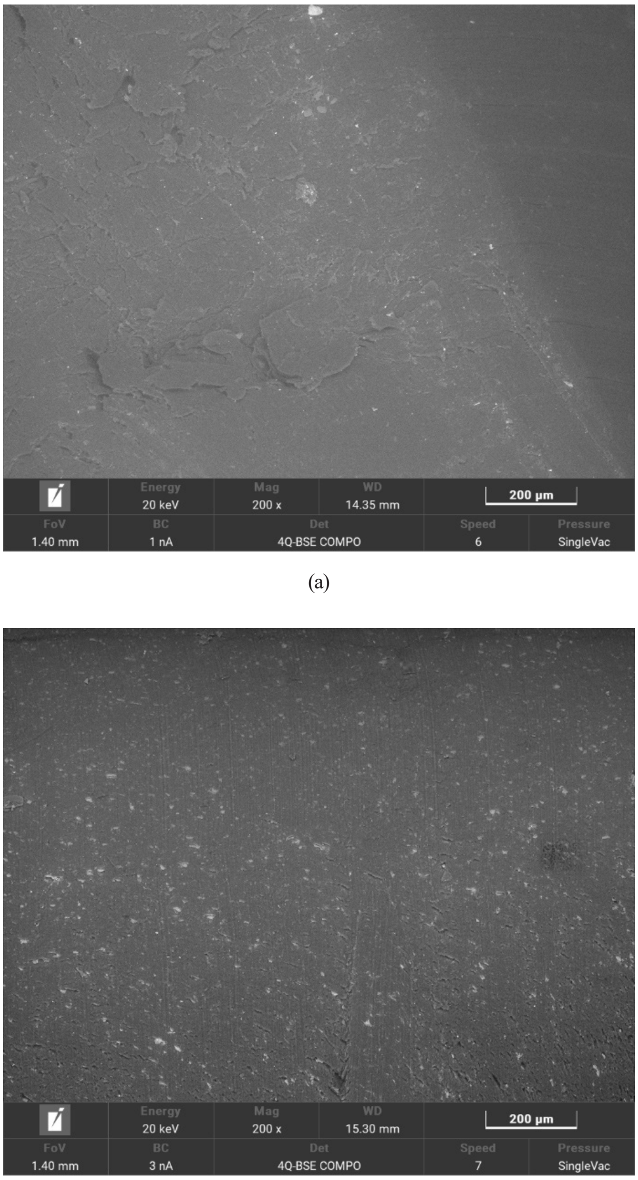
To investigate the degradation behavior and the changes in the thermal properties of recycled PLA and PLA-IW compounds, several DSC measurements were conducted. The names of the recycled samples are PLA_R_X, where

Table 1 Injection molding parameters for sample preparation

Processing parameters	Values for PLA
Dosing volume (cm^3)	84
Injection speed (cm^3/s)	100-60 (2 phase)
Injection pressure (bar)	1200
Holding pressure (bar)	1000
Holding pressure time (s)	15
Barrel temperatures (°C)	195-175
Back pressure (bar)	80
Mold temperature (°C)	25

Table 2 IW, PLA, and PBAT content of the produced compounds

IW content (w%)	Recycled PLA content (w%)	PLA content (w%)	PBAT content (w%)
0	100	100	0
10	90	98.5	1.5
30	70	95.5	4.5
50	50	92.5	7.5
0	30	89.5	10.5
90	10	86.5	13.5
100	0	85.0	15.0



(a)

(b)

Fig. 1 SEM micrographs of compounds containing (a) 10 w% IW, and (b) 50 w% IW

X indicates the number of processing cycles that the material went through. As for the IW containing samples all labels refer to the IW content in w% form.

Crystallinity was calculated from melt enthalpies and the equilibrium melting enthalpy of PLA (93.1 J/g) reported by Saeidlou et al. [26]. According to the results of neat PLA (Table 3), the glass transition temperature slightly decreased from 59.49 °C to 55.94 °C with 10 rounds of injection molding. Badia and Ribes-Greus [27] also obtained similar results when investigating the mechanical recycling of PLA. In their case, the decrease in the glass transition temperature was associated with the increase in free volume and the decrease in molecular weight.

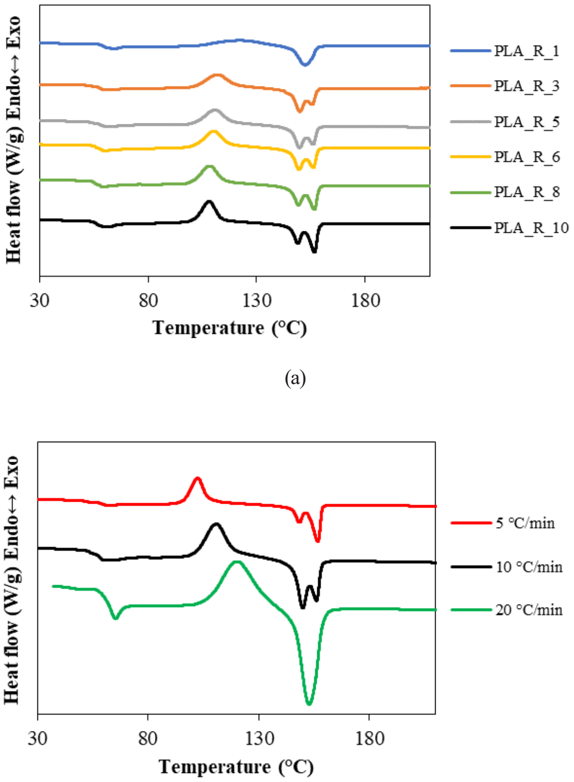
All samples exhibited cold crystallization during heating, so the crystallinity of the injection molded samples was calculated from the difference between the cold crystallization and melting enthalpies. Due to the inaccuracies in the process of evaluation, there is no clear trend observable; however, it can be stated that the average crystallinity of the injection molded samples is around 5 %. Peaks associated with cold crystallization and melting both increased with each recycling step, indicating that PLA's ability to crystallize improved. The increasing crystallinity with the number of recycling cycles corresponds to the results obtained by Zenkiewicz et al. [19]. The rising crystallinity was attributed to the potential nucleating effect of degradation by-products or impurities, but lower molecular weight of the degraded material can also play a part.

Fig. 2(a) shows from the third round of recycling two melting peaks were visible, one at 149 and another at 156 °C. The area of the second peak grew with the number of recycling processes while the first one decreased. To investigate whether the two peaks were a result of different crystalline forms in the samples or the occurrence was due to the rearrangement of the structures at high temperatures, measurements with 5 °C/min and 20 °C/min heating rates were performed. Samples from PLA_R_1, PLA_R_5, and PLA_R_10 were investigated further and t showed similar tendencies. The results of PLA_R_5 are shown in Fig. 2(b). At 5 °C/min, the area of the second peak was larger, while at 10 °C/min it was smaller. At 20 °C/min heating rates, the two peaks could not be separated.

The change between the ratios of the peaks implies that only one crystalline form is present, and at slower heating rates exothermic recrystallization was possible due to the existence of both thermodynamical and kinetic conditions. Considering the possible crystalline forms of PLA,

Table 3 Thermal properties of recycled PLA and PLA-IW compounds

Sample name	Glass transition temperature (°C)	Crystallization temperature (°C)	Crystallization enthalpy (J/g)	Melting enthalpy (J/g)	Crystallinity (%)	Crystallinity after cold crystallization (%)
PLA_R_0	-	-	-	33	35	35
PLA_R_1	59.5	122.2	18.4	22	4	21
PLA_R_2	58.0	120.7	23.2	27	5	25
PLA_R_3	58.0	112.3	25.7	32	7	30
PLA_R_4	58.2	111.1	27.0	30	4	28
PLA_R_5	58.3	110.4	26.7	32	6	30
PLA_R_6	58.5	109.5	29.9	32	2	30
PLA_R_7	55.4	108.5	28.1	34	6	32
PLA_R_8	55.5	108.6	28.1	35	7	33
PLA_R_9	55.2	108.2	28.4	36	8	33
PLA_R_10	55.9	108.3	30.1	34	4	32
100 w% IW	55.5	108.4	5.5	7	2	7
90 w% IW	59.3	106.1	8.1	10	2	10
70 w% IW	56.4	107.6	9.1	14	4	14
50 w% IW	55.1	107.1	15.0	20	5	19
30 w% IW	56.0	109.7	16.5	21	5	21
10 w% IW	55.8	108.3	21.1	27	6	27



(a)

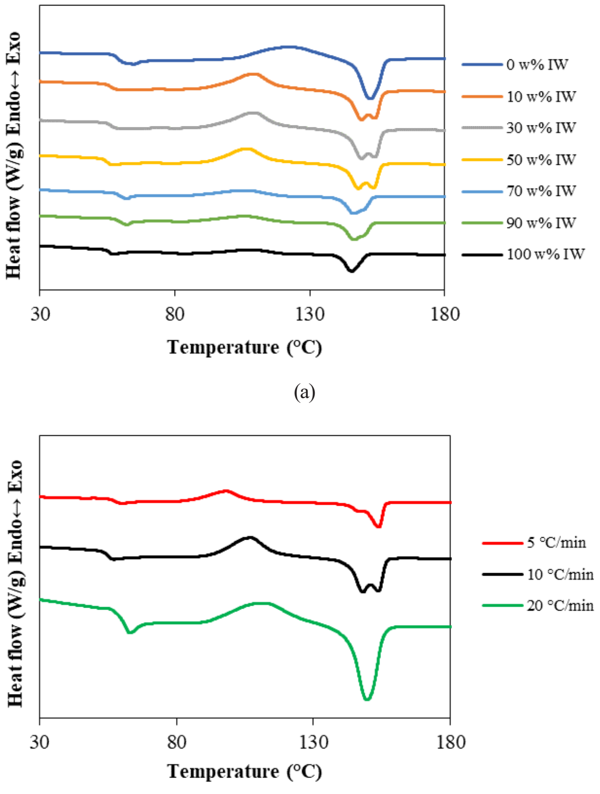
(b)

Fig. 2 (a) DSC curves of recycled PLA, and (b) PLA_R_5 at different heating rates

the two peaks probably belong to the less ordered *α*' form and its change to the more ordered *α* form [28]. During injection molding, the mold temperature was 25 °C which caused the polymer melt to cool instantly, resulting in a less ordered crystalline structure.

Fig. 3(a) shows the variations of the DSC curves for samples with different mass ratios of PLA and IW. The glass transition temperature, as a characteristic parameter, is approximately in the same temperature range on every curve, which is around the Tg of PLA (55 to 60 °C). PBATs Tg was measured at 29.6 °C during separate measurements that were conducted in a wider temperature range just to show that it can be detected in the compounds. The minimal change in glass transition temperature with increasing PBAT concentration confirms that PBAT does not blend with PLA.

The area under the peak and the maximum temperature associated with cold crystallization decreased with decreasing PLA content changing from 122 °C to around 108.5 °C when PBAT was present. However, for the sample containing 100 % PLA, the peak is notably smaller. According to the work of La Mantia et al. [25] used in small amounts, PBAT can act as a nucleating agent, resulting in higher crystallinity when only 10 w% IW was used. In compounds that contain more IW, PBAT hinders crystallization. The melting peak is more important



(a)

(b)

Fig. 3 (a) DSC curves of PLA and IW compounds, and (b) 50 w% IW at different heating rates

as it provides precise information about the structural and thermal properties of the sample and the complexity of the melting process. As shown in Fig. 3 and Table 3, the melting temperature, the melting enthalpy, and thus the crystallinity gradually increases with increasing PLA content. The phenomenon of double peaks can be observed in some compositions. This could be due to the phenomenon of crystalline perfection upon heating, or, since we investigated a two-component system, polymorphism might also occur.

To investigate the double peak phenomenon more thoroughly, DSC measurements were conducted at various heating rates because the proportion of modifications in the system is dependent on the speed of heating [29, 30].

In Fig. 3(b), it can be seen that at a heating rate of 20 °C/min, the 50 w% IW compound does not exhibit double peaks. In contrast, at 5 °C/min heating rate, the double peak phenomenon is present in the curves, and there is also a change in the melting enthalpy (Table 3). With increasing heating rate, the peak at higher temperatures shifts towards lower temperatures and decreases in size. Therefore, as it can be observed in Fig. 3(b), the double peak phenomenon disappears at a heating rate of 20 °C/min. This can be explained the same way as with neat PLA, where recrystallization was possible at 5 °C/min heating rate, but not at 20 °C/min.

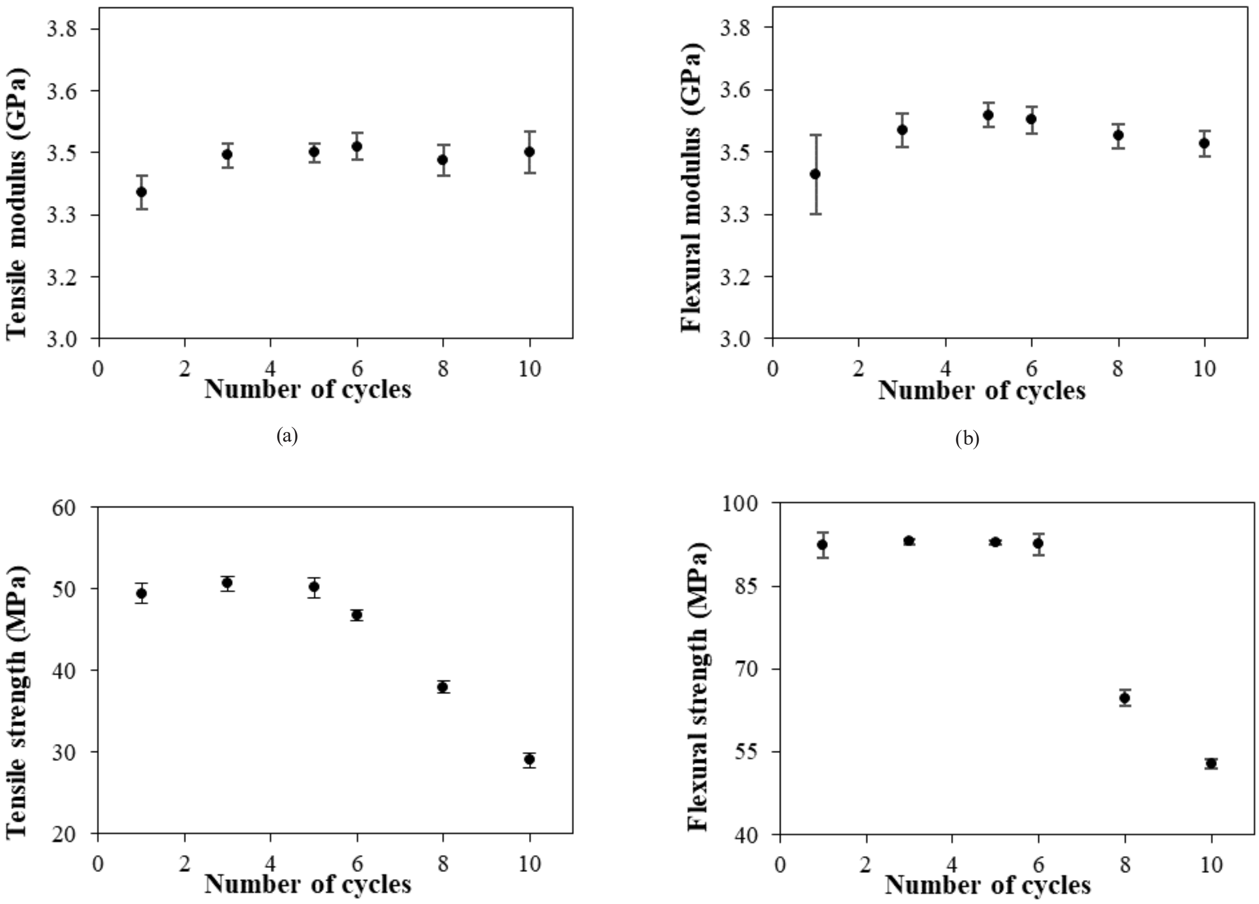
3.3 Mechanical properties

The changes in the mechanical properties of recycled PLA are shown and Table 4. The tensile tests showed that the tensile and flexural modulus of PLA do not change significantly with recycling (Figs. 4(a) and 4(b)). The tensile strength (Fig. 4(c)) did not change for 6 cycles and started to drop with each new cycle and similar tendencies were observed with the flexural strength (Fig. 4(d)). After the 10th cycle, the tensile strength was 27.94 MPa, which is a 45% decrease from the single-time injection molded samples.

The deformation at break changed similarly from 2.42 to 0.84% with tensile testing, and from 6.48 to 1.51%

Table 4 Mechanical properties of recycled PLA and PLA/IW compounds

	Tensile modulus (GPa)	Tensile strength (MPa)	Deformation at break (%)	Flexural modulus (GPa)	Flexural strength (MPa)	Deformation at break (%)
PLA_R_1	3.35	51.73	2.42	3.40	92.34	6.48
PLA_R_3	3.44	49.63	2.41	3.50	93.06	6.34
PLA_R_5	3.45	49.71	2.06	3.54	92.91	4.78
PLA_R_6	3.47	47.38	1.51	3.53	92.54	3.54
PLA_R_8	3.43	37.32	1.15	3.49	64.70	1.86
PLA_R_10	3.45	27.94	0.84	3.47	52.77	1.51
10 w% IW	3.68	50.68	1.74	3.58	90.09	4.18
30 w% IW	3.38	45.73	1.94	3.33	79.57	3.85
50 w% IW	3.35	40.98	1.99	3.28	70.15	3.45
70 w% IW	2.94	34.12	1.98	2.83	58.20	3.56
90 w% IW	2.13	23.90	3.81	2.05	42.35	4.56
100 w% IW	1.77	20.57	8.57	1.65	36.70	5.15



(a) (b)

(c) (d)

Fig. 4 (a) Tensile modulus, (b) flexural modulus, (c) tensile strength, and (d) flexural strength of the recycled PLA

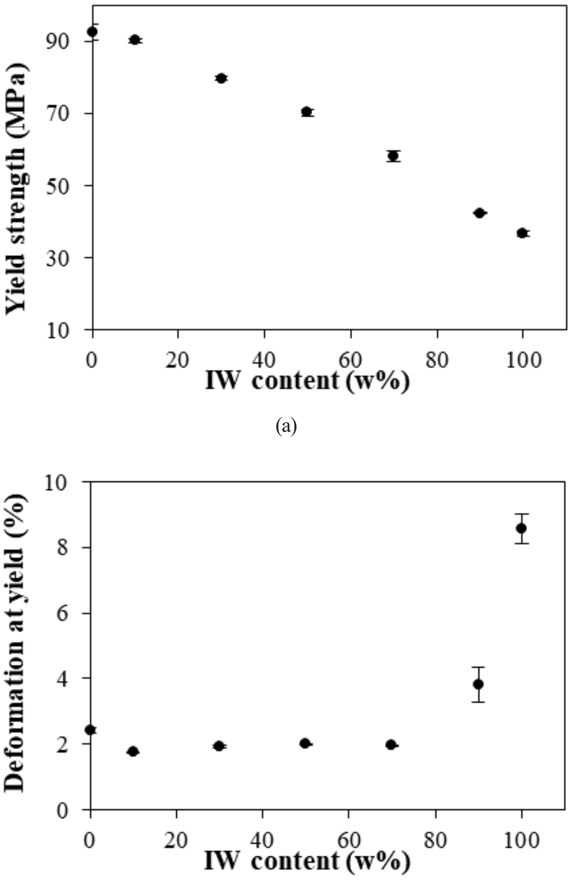
during bending tests. All in all, mechanically recycled PLA had very small changes in mechanical properties for up to 6 cycles of processing and deteriorated harshly after. This was not following the findings of Zenkiewicz et al. [19]. In their research, it was found that the tensile properties of PLA do not change significantly with the number of processing cycles, specifically extrusion. However, measurements conducted by them had different conditions (crosshead speed and sample dimensions) which can be accountable for different results.

PBAT has been widely investigated as a toughening agent for biodegradable polymer blends [31, 32]. Post-consumer plastic waste can contain a lot of different polymers, so if biopolymers were to be collected separately depending on the concentration of PBAT (and other types of biopolymers), the recycled material's properties could vary significantly. This is what we aimed to investigate further in this research.

As expected, the modulus and yield strength were higher when the compounds contained less IW (Fig. 5(a)). The industrial waste caused a 51% decrease in the tensile modulus when it was recycled on its own.

This was due to the PBAT's ability to make the compound more flexible and less stiff, which was well represented in the deformation values of the materials. At 1 mm/min crosshead speeds the samples composed of 90 w% and 100 w% industrial waste did not brake during the test, therefore only yield stress and deformation at yield were evaluated. Deformation at yield highly increased with IW content, being at its maximum at 8.6 % with the neat IW samples. Our assumptions proved to be valid, with the recycling of different types of biopolymers together. The mechanical properties change drastically even with less than 15 w% of PBAT content.

The impact strength slightly decreased with the number of cycles (Fig. 6(a)), although the results showed high deviations. After three rounds of injection molding, the impact strength did not change, and similar to the tensile and bending properties it started to decrease with the 5th cycle, indicating that PLA is fully recyclable for at least 3-4 times. It is important to note, that the impact strength of PLA was low to begin with at 1.75 kJ/m^2 and changed to 1.25 kJ/m^2 after 10 cycles, which is a 28.6% decrease.



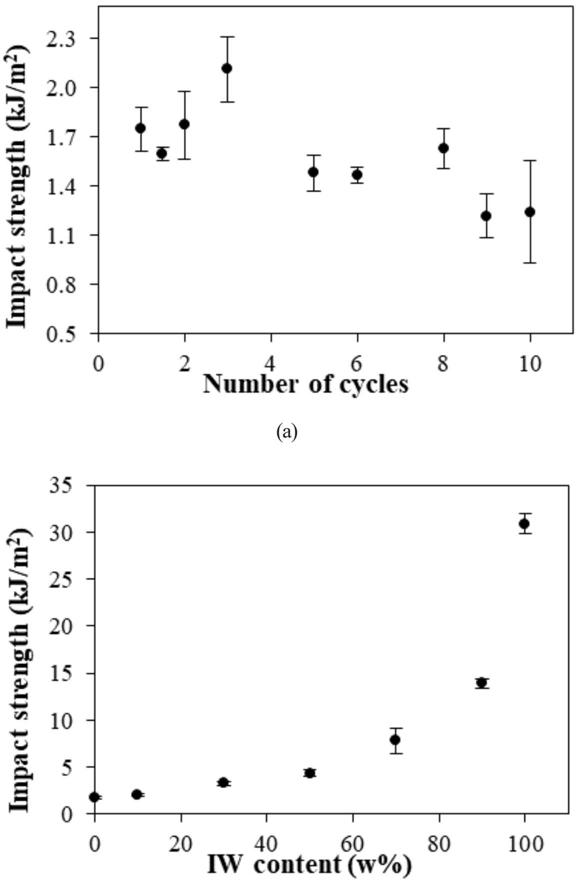
(a)

(b)

Fig. 5 (a) Yield strength, and (b) deformation at yield of PLA and IW compounds

Different tendencies were observed for the samples containing different ratios of industrial waste (Fig. 6(b)). The impact strength of the PBAT containing industrial waste was 31 kJ/m^2, almost 18 times more than the pure PLA sample. PBAT is generally used to modify the impact strength of PLA [32] so recycling the two polymers together results in different mechanical properties depending on the ratio of the components. All mechanical tests combined indicate that the presence of PBAT increased the impact strength but decreased the tensile modulus and tensile strength.

The heat deflection temperature showed a steady decrease with the number of recycling cycles, ranging from 58.3 °C for neat PLA and 57.3 °C for 10 times processed PLA (Fig. 7(a)). This small difference correlates with the decrease in the glass transition temperature examined by DSC. Similar conclusions can be drawn about the recycled industrial waste compounds. Fig. 7(b) shows that with increasing IW content the heat deflection temperature decreased, being the highest of neat PLA and the lowest at 90 w% of IW content and 56.1 °C.



(a)

(b)

Fig. 6 Impact strength as (a) a function of processing cycles, and (b) industrial waste content

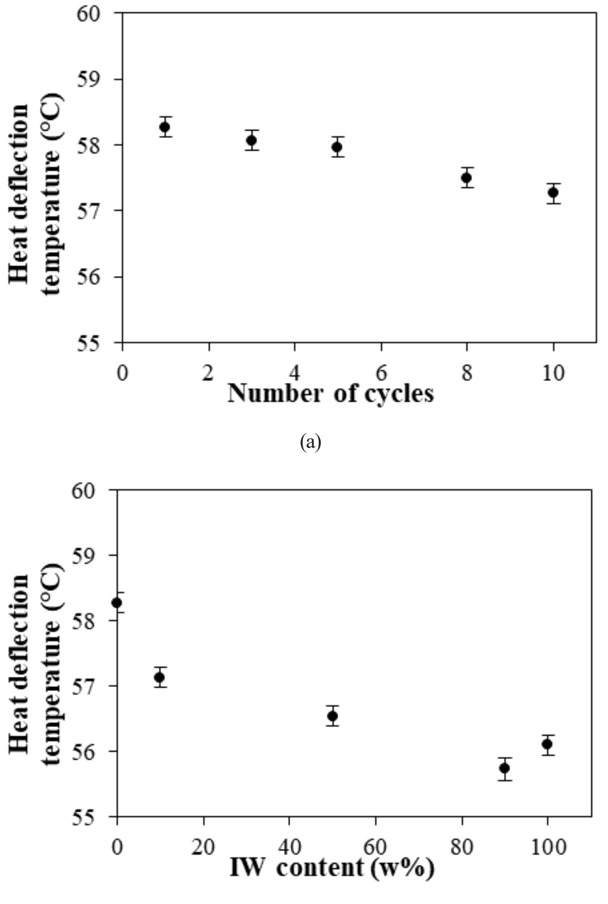
3.4 Rheology

The MFR values increased drastically with recycling. After one round of processing PLAs MFR increased from 18.05 to 35.48 g/10 min (Fig. 8(a)). Samples that were processed 8-10 times, could not be measured properly with this technique. The growth of MFR means a decrease in viscosity, which can be associated with degradation and decrease in molecular weight caused by multiple processing cycles [33]. The lower viscosity and molecular weight could also play a part in the deterioration of the mechanical properties after multiple recycling rounds.

In the case of PLA and IW blends (Fig. 8(b)), the changes in the MFR values can be mostly attributed to the difference of the neat materials MFRs, which were 34.4 g/10 min for PLA processed for 1 time, and 13.5 for the recycled PBAT containing IW.

4 Conclusions

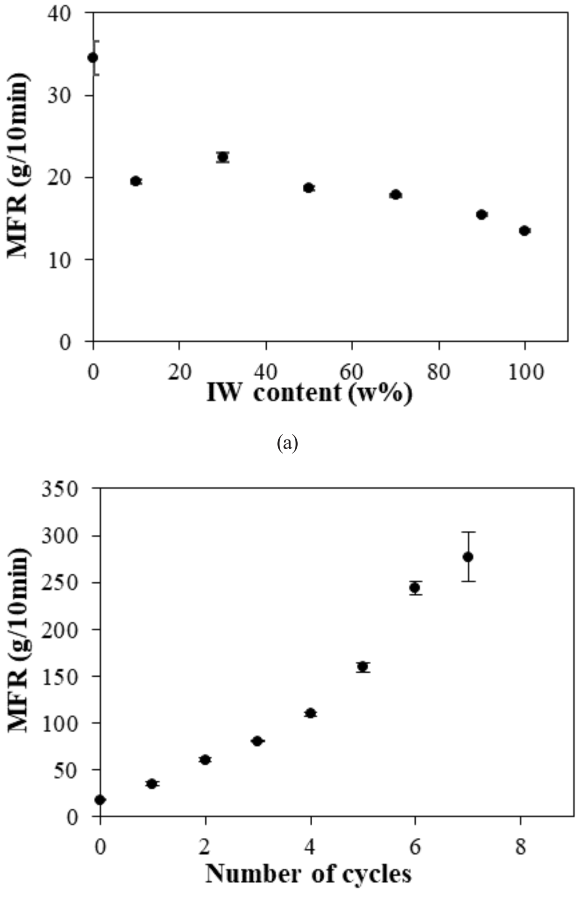
During the series of experiments investigating the mechanical recyclability of PLA, it was observed that PLA undergoes degradation upon multiple recycling cycles. Its processability



(a)

(b)

Fig. 7 Heat deflection temperature values as a (a) function of processing cycles, and (b) industrial waste content



(a)

(b)

Fig. 8 MFR values as (a) a function of processing cycles, and (b) industrial waste content

becomes increasingly difficult, and process parameters need to be adjusted to manufacture samples. This is supported by the MFR measurements as the values increased with the number of processing cycles: 18 g/10 min after the first cycle and 277 g/10 min after the seventh cycle. For more than 7 cycles of processing, MFR could not be measured under the applied conditions. The decrease in molecular weight due to degradation was further confirmed by the increasing crystallization tendency observed in the DSC analysis. It is important to highlight that from both technological and material standpoints, processing for 5-6 cycles proved to be achievable. After that, mechanical properties start to deteriorate, with decreasing tensile strength, deformation, and impact resistance. This finding contradicts what was found in the literature, where PLA's properties were seemingly unchanged after 10 times of extrusion [19]. Heat deflection temperature and melt viscosity also decrease with the number of processing cycles.

Our experiments showed that PLA and PBAT products can be recycled together, even if they originally required different processing conditions. However, the properties of the material depend highly on the ratios of PLA and PBAT. According to the DSC results, the amount of crystalline fraction increases with the increase in pure PLA content, as PBAT's random copolymer structure inhibits significant crystallization. However, even with a small amount of PBAT content, the crystalline fraction increased, and based on MFR results, viscosity also increased, which can be explained by PBAT acting as a nucleating agent. Variable heating rate DSC measurements were conducted to examine the phenomenon of double peaks observed during heating. The results indicate crystalline perfection occurring during heating, presenting as a secondary peak within the melting peak.

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