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Abstract

Poly(lactide) (PLA) is the most versatile biopolymer, but it has an unfavorable end-of-life. Biodegradation occurs under strictly defined conditions, and ultimately, it is landfilled, where it behaves like conventional plastics. To completely utilize the potential of PLA, it is necessary to rethink end-of-life scenarios. In this work, the influence of 10 cycles of reprocessing PLA pre-consumer industrial waste on the material's properties was examined. The mechanical milling of the material was followed by injection molding, and after each cycle, mechanical, thermal, chemical, and optical properties were studied. Between the cycles, no virgin PLA or any additives were added to enhance the properties. Results showed a slight decrease in molecular weight, while the thermal properties remained unchanged compared to the starting material.

Keywords: bioplastics; poly(lactide); industrial waste; mechanical recycling; thermal properties; mechanical properties

1. Introduction

The growing demand for environmentally sustainable materials has driven the increasing adoption of bio-based polymers, among which poly(lactide) (PLA) has emerged as a leading alternative to petrochemical plastics. Derived from renewable resources such as corn starch and sugarcane, PLA offers advantages including biodegradability, biocompatibility, and favorable mechanical and thermal properties, making it widely used in packaging, textiles, 3D printing, and biomedical applications [1–4].

Despite its ecological appeal, the environmental benefit of PLA depends largely on how it is managed at the end-of-life (EoL). PLA is compostable under industrial conditions (≥58 °C, high humidity, microbial activity), yet, its biodegradation in natural or home composting environments is extremely slow, often requiring months to years [5,6]. In practice, a significant share of PLA ends up in landfills or incineration, undermining its circular potential [7]. Therefore, viable recycling strategies are critical for realizing the full sustainability of PLA.

Several EoL routes are available for PLA, including mechanical recycling, chemical depolymerization, industrial composting, and incineration with energy recovery [8–17]. Among these, mechanical recycling is recognized as the most economically and energetically feasible pathway in the short term, as it allows for the reuse of the polymer without depolymerization, maintaining the material's structural integrity through physical reprocessing steps such as shredding, melting, and extrusion [18,19]. Mechanical recycling also aligns well with circular economy goals, especially in industrial and post-consumer contexts such as 3D printing and food packaging.

However, repeated mechanical recycling of PLA might introduce thermal and hydrolytic degradation, primarily through chain scission, leading to reduced molecular weight, diminished

mechanical properties, and altered viscosity [20]. These limitations restrict the recyclability of PLA, particularly for applications demanding high mechanical performance.

To mitigate such degradation, researchers have developed various strategies:

- Chain extenders, including epoxy-functionalized additives like Joncryl® ADR, have been shown to restore molecular weight, increase viscosity, and stabilize mechanical properties over multiple cycles [21,22].

- Blending recycled PLA with virgin material (typically 30–70%) maintains mechanical integrity while enhancing sustainability [23].

In this work, pre-consumer waste underwent 10 processing cycles and properties were studied after each to better understand the influence of reprocessing on mechanical, thermal, chemical and optical properties of PLA compared to the starting material.

2. Materials and Methods

Stramex PET d.o.o. (Podplat, Slovenia) supplied PLA bottles (pre-consumer industrial waste). They were used as received, without any treatment.

2.1. Processing of the Material

Bottles were milled using Wanner milling machine for thermoplastics extruded on the twin-screw extruder Labtech LTE 20-44. The screws diameters 20 mm, L/D ratio 44:1, screws rotation 600 rpm, temperature profile was from the hopper (165 °C) to the die (180 °C).

After extrusion, PLA pellets were processed using ARBURG injection molding machine (Allrounder 320 C500-100 Golden Edition). The temperature was increasing from the hopper (165 °C) to the die (180 °C). The tool temperature was set to 45 °C and the cooling time to 20 s. After the first cycle of injection molding, molded samples were milled and molded in nine more consecutive cycles. From each cycle, 10 samples for further examination were taken. Between the cycles, milled material was dried at 75 °C until the moisture content was below 0.025 % which was determined using the Mettler Toledo HX204 moisture analyser. No plasticizer or any other additive was added to PLA during the recycling.

2.2. Characterization of the Material

The melt flow index (MFI) of samples was evaluated according to ISO 1133 via MFI indexer (Dongguan Liyi Test Equipment, type LY-RR) at a temperature of 210 °C with a 2.16 kg weight. Results are expressed as average value with standard deviation.

Thermal measurements were carried out with a differential scanning calorimeter (DSC 2, Mettler Toledo) under a nitrogen atmosphere (20 mL/min). The sample temperature was increased from 0 to 200 °C at a heating rate of 10 °C/min and held in the molten state for 5 min to erase their thermal history. After being cooled at 10 °C/min, the samples were reheated to 200 °C at 10 °C/min. The crystallization temperature (Tc), crystallization enthalpy (ΔHc), melting temperature (Tm) and melting enthalpy (ΔHm) were obtained from the cooling and the second heating scan. Moreover, the relative crystallinity values (Xc) were calculated, using following equation:

$$Xc\ (\%) = \frac{\Delta Hm - \Delta Hcc}{\Delta Hf} x100,$$

where ΔHm refers to enthalpy of melting

ΔHcc refers to enthaly of cold crystallization

ΔHf refers to enthalpy of melting of 100% crystaline PLA and equals 93,6 kJ/g

Thermogravimetric analyses (TGA) were performed on a Mettler Toledo TGA/DSC3. The analyses were carried out in a nitrogen atmosphere (20 mL/min) from 40 to 600 °C with a heating rate of 10 °C/min, followed by a segment in the oxygen atmosphere (20 mL/min) from 600 °C to 700 °C while the heating rate remained the same.

Thermo-mechanical properties were examined using a Perkin Elmer DMA 8000. The samples were heated at 2 °C/min from room temperature to 170 °C under an air atmosphere. A frequency of 1 Hz and an amplitude of 20 μm were used in dual cantilever mode.

Impact tests were performed on the pendulum Dongguan Liyi Test Equipment, type LY-XJJD5, according to ISO 179. The distance between supports was 60 mm, 2 J pendulum was used.

Flexural and tensile tests were conducted on the Shimadzu AG-X plus according to ISO 178 and ISO 527-1, respectively. Five measurements were performed for each sample.

Chemical properties were examined via infrared spectroscopy with Fourier transformation (FTIR, IRAffinity-1S, Shimadzu), measuring the transmittance (T%), in the wavenumber range 4000-400 cm^{-1} and 4 cm^{-1} resolution.

Crystallinity was determined using an X-ray diffractometer, Rigaku MiniFlex 600 (XRD), using Cu-Kα radiation. The 2θ range was from 0 to 50, resolution 0.04, and 3 s hold.

Surface properties of materials were determined by measuring contact angle against water (Osilla Goniometer). One drop of distilled water was placed on the surface of the material, and the contact angle was determined using software.

The colorimetric properties of the samples, subjected to multiple recycling cycles, were evaluated using the CIELAB color space system, comprising parameters L*, a*, b*, C*, and h*. These parameters provide a comprehensive representation of the material's perceived color. L* denotes lightness (0 for black, 100 for white), a* represents the red-green axis (positive for red, negative for green), b* represents the yellow-blue axis (positive for yellow, negative for blue), C* signifies chroma or saturation, and h* indicates the hue angle. A 3nh NR60CP spectrophotometer was used for evaluation of described parameters.

3. Results and Discussion

Figure 1 illustrates changes in MFI and viscosity with consecutive processing of PLA. Except for cycles 2 and 3 (samples PLA2 and PLA 3), MFI is increasing, while the viscosity of the melt is decreasing. Since no processing additives were added to the PLA, these changes are a direct consequence of consecutive milling and injection molding, which induced a decrease in molecular weight of the PLA molecules.
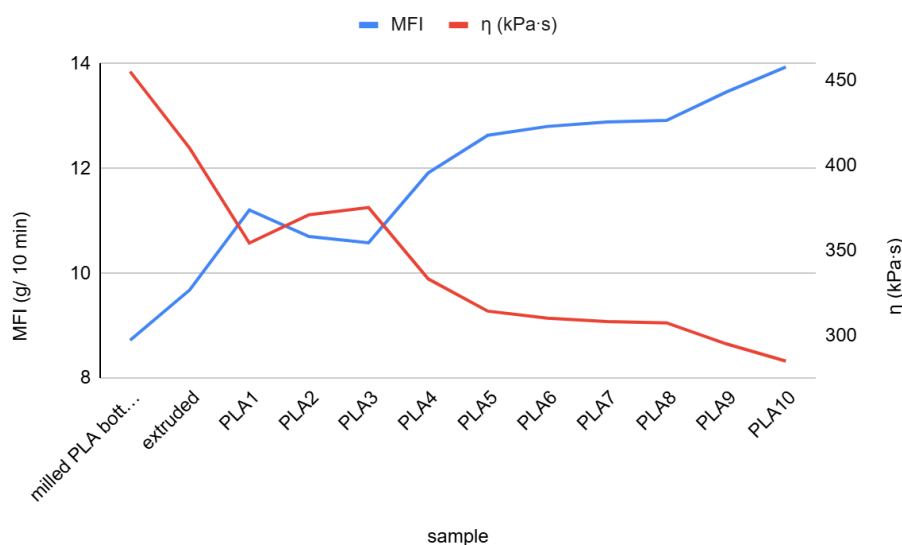

Figure 1. MFI (blue line) and viscosity (red line) of recycled PLA.

Thermal properties of the examined samples are summarized in Table 1 and the thermograms of second healting cycle are given at Figure 2. The present results are from the second heating cycle.

It can be seen that there are no changes in Tg, Tc and Tm, they are very constant with low deviation from first to last cycle. There are changes in the values of both crystallization and melting enthalpies, which don't follow the regular trend of change. Variations in enthalpies might be a consequence of different crystalization time of the samples, since the processing was not done the same day for all cycles.

Table 1. Thermal data from 2nd heating cycle.

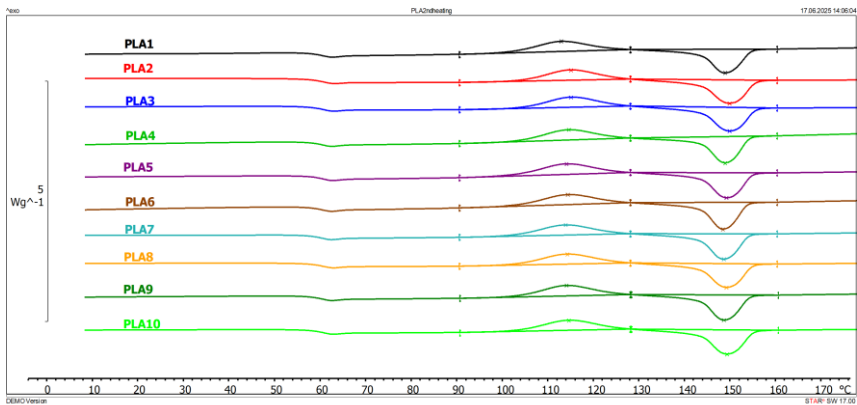| Sample	Tg (°C)	Tcc (°C)	ΔHcc (J/g)	Tm (°C)	ΔHm (J/g)	Xc (%)	Td$_1$ (°C)
Milled PLA	59.5	113.1	17.6	148.9	26	8.96	370.1
Extruded PLA	59.3	114.4	22.21	148.8	23.21	1.07	369
PLA1	59.2	114.4	23.2	148.7	24.09	0.95	368.7
PLA2	59.9	115.2	16.79	149.7	28.84	12.86	368.5
PLA3	59.7	115.2	16.35	149.8	28.69	13.17	368.1
PLA4	59.5	114.7	16.93	148.9	28.25	12.08	367.5
PLA5	59.5	115	24.47	149.6	25.53	1.13	366.9
PLA6	59.7	114.2	18.73	149.2	28.36	10.28	367.1
PLA7	59.4	114.5	17.24	148.4	27.94	11.42	366.6
PLA8	59.4	114	17.74	148.5	27.59	10.51	364.8
PLA9	59.7	114.4	17.3	149.2	27.86	11.27	365.3
PLA10	59.5	114.9	23.15	149.4	25.16	2.15	363.7

Figure 2. Thermograms of the second heating cycle.

The degradation temperature (Td$_1$) was determined by the TGA method. The values have an almost linear trend of decrease, from 370 °C to 363.7 °C (Table 1), which is in line with MFI results. When MFI is higher, molecular weight is lower, and it is expected that within corresponding samples Td$_1$ will be lower. Even though the degradation temperature is decreasing, the maximum difference is below 7 °C, which is only 2%. Considering DSC and TGA data, it can be concluded that thermal stability did not significantly change with multiple consecutive processing of PLA.

Dynamic-mechanical analysis results give insight into stiffness, storage modulus, and mechanical Tg values, which reflect mechanical softening (segmental motion). The difference in Tg values is low, less than 1 °C, which is in accordance with DSC results. The difference in Tg values determined by DSC and DMA (Table 2 and Figure 3) is expected, since the methods deliver different types of Tg. DSC is the best for thermodynamic Tg, which is related to the processing and the thermal stability of the material, while DMA gives insight into mechanical Tg, which is related to the functional behavior and the mechanical material's design. DMA Tg has a higher value, and the expected difference compared to DSC Tg is from 5 to 20 °C, which is proven in this case, where DMA Tg has a higher value for 6 to 7 °C. Samples PLA5, PLA6, and PLA8 have two, and PLA7 has three crystallization peaks, which refers to stepwise or incomplete crystallization. It can also be the result

of heterogeneous nucleation or processing of the material on different days and different crystallization time for the material.

Table 2. Results of DMA analysis of PLA samples.

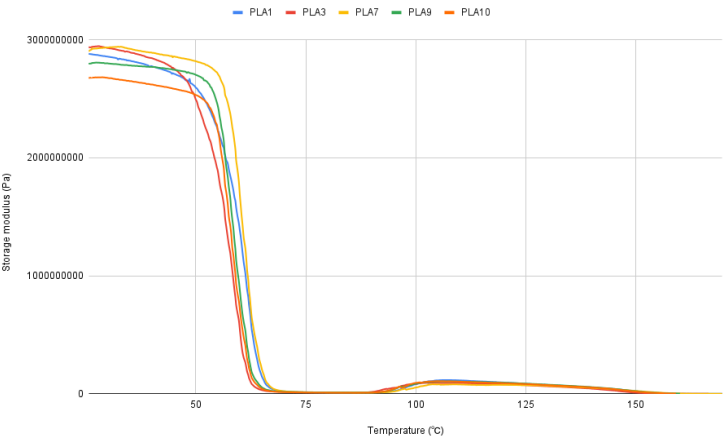| Sample	E' @ 30 °C (GPa)	tanδ	Tg	tanδ cc (-)	tanδ cc (°C)	E' cc (GPa)
PLA1	2.8561	1.942	66.96	95,1	0,168	0,11
PLA2	2.949	1.903	67.28	95,3	0,156	0,10
PLA3	2.9365	1.966	67.56	95,7	0,166	0,09
PLA4	3.0025	1.988	67.48	94,5	0,162	0,10
PLA5	2.7808	1.973	67.44	95,6/107,7	0,171/0,126	0,07/0,08
PLA6	2.8575	1.928	66.64	95,1/109,2	0,162/0,090	0,10/0,09
PLA7	2.9219	1.968	66.72	95,6/105,2/115,3	0,158/0,122/0,106	0,08/0,07
PLA8	3.0066	1.991	66.76	95,4/106,6	0,159/0,135	0,07/0,08
PLA9	2.8005	1.961	66.4	93,3	0,151	0,10
PLA10	2.6755	2.049	67.19	94,3	0,166	0,10

Figure 3. DMA curves of selected samples.

The reaction of the material to high stress load can be measured by an impact test, resulting in the impact strength and evaluation of brittleness/ductility of the material. Brittle materials, like PLA, have very low impact strength in both notched and unnotched examined samples. Comparing the impact strength from the first to the tenth cycle within notched samples, the value slightly decreases from 2.85 to 2.55 kJ/m² (Figure 3). Within unnotched samples, sample PLA8 has the highest impact strength of 22.3 kJ/m², while PLA3 sample has the lowest impact strength of 14 kJ/m². All samples, notched and unnotched, have low energy absorption potential and have fragile break behavior with almost no plastic deformation.

Flexural and tensile properties of the material are of the highest importance when it comes to the end-use application. Recycled materials usually do not meet requirements for high-performance use, so they are used for more technical applications. In the case of multiple recycling of PLA, it can be seen (Table 3) that (considering standard deviation), there are no significant differences between flexural strength and strain at strength of the examined samples. This indicates that samples retain rigidity, even after ten cycles of thermal processing.

Table 3. Flexural test results.

Sample	Flexural Modulus (GPa)	Flexural Strength (MPa)	Strain at Strength (%)
PLA1	3.25 ± 0.01	94.41 ± 0.05	4.25 ± 0.05
PLA2	3.23 ± 0.05	94.19 ± 0.25	4.24 ± 0.02

PLA3	3.3 ± 0.07	94.5 ± 0.63	4.29 ± 0.09
PLA4	3.28 ± 0.08	98.59 ± 0.32	4.35 ± 0.02
PLA5	3.32 ± 0.03	93.87 ± 0.84	4.1 ± 0.17
PLA6	3.31 ± 0.01	93.78 ± 0.44	4.21 ± 0.04
PLA7	3.33 ± 0.03	94.59 ± 0.43	4.14 ± 0.13
PLA8	3.34 ± 0.05	95.27 ± 0.47	4.25 ± 0.05
PLA9	3.44 ± 0.03	97.31 ± 1.58	4.08 ± 0.4
PLA10	3.39 ± 0.05	98.05 ± 0.26	4.3 ± 0.06

Tensile test results are summarized in Table 4. Measured values indicate that the material is brittle and rigid (which is also proven with previous DMA and impact test results) and has very small elongation before break. Differences in tensile strength and strain at break are low and are not significant, and it can be concluded that multiple recycling does not affect the tensile properties of PLA. Comparing the values of the flexural and tensile tests, it can be concluded that the material can withstand higher stress loads in the transversal direction than in the longitudinal direction, since a higher force is necessary for the material to break.

Table 4. Tensile test results.

Sample	Tensile modulus (GPa)	Tensile Strength (MPa)	Strain at Strength (%)	Strain at Break (%)
PLA1	2.62 ± 0.23	68.2 ± 2.72	4.58 ± 0.29	5.18 ± 0.78
PLA2	2.55 ± 0.26	67.4 ± 0.51	4.48 ± 0.36	5.72 ± 0.55
PLA3	2.66 ± 0.33	67.9 ± 1.16	4.36 ± 0.35	5.34 ± 0.61
PLA4	2 ± 0.25	69 ± 0.73	4.22 ± 0.16	5.92 ± 0.9
PLA5	3.18 ± 0.15	67.5 ± 0.85	4.15 ± 0.24	4.94 ± 0.49
PLA6	2.69 ± 0.38	66.9 ± 0.92	4.08 ± 0.15	5.79 ± 0.64
PLA7	3.21 ± 0.32	66.6 ± 0.71	4.13 ± 0.15	4.99 ± 0.16
PLA8	2.73 ± 0.15	65.5 ± 0.74	4.13 ± 0.08	5.04 ± 0.37
PLA9	2.6 ± 0.33	66.3 ± 0.34	4.19 ± 0.15	5.67 ± 0.62
PLA10	2.86 ± 0.23	67.1 ± 0.8	4.09 ± 0.09	5.3 ± 0.41

Changes in the chemical structure of PLA during multiple processing are determined by the FTIR method, and spectra are illustrated in Figures 3 and 4. The difference in peaks around 3000 cm^{-1} was detected, while the difference around 2360-2320 cm^{-1} is related to the CO_2. Selection of peaks from 3100 to 2800 cm^{-1} is shown in Figure 4. Spectra of PLA1 and PLA3 (bottom blue and yellow lines) have a different shape compared to the others. Spectra of samples PLA9 and PLA10 do not have the peak at 2850 cm^{-1}. Peaks from 2995 to 2945 cm^{-1} are related to the CH stretching, the peak at 2879 cm^{-1} is related to asymmetric -CH$_3$ stretching, while the peak at 2852 cm^{-1} corresponds to the symmetric -CH$_3$ or -CH$_2$ stretching. The difference in those peaks can indicate the polymer chain scision, which is in correlation with MFI results.

According to the literature [24], PLA has a contact angle between 70 and 80 °, which makes its surface moderately hydrophilic to slightly hydrophobic. Values obtained during this experiment fall into this range (Figure 5), except for samples PLA2 (lower value - more hydrophilic), PLA4 and PLA9 (higher value - more hydrophobic). Images of water drops on the sample surface are illustrated in Figure 6. This can also be connected to the different relaxation times for different series of samples, which can cause the difference in surface properties and orientation of the groups in the polymer chain.

Crystallinity of selected samples is shown in Figure 7. It can be seen that there is no change in the crystallinity of samples within different cycles. A wide peak from 10 to 25 °, with a maximum at 16.7 °, which is according to literature data [25], is a result of the scattering of PLA. One peak with very low intensity occurs around 34 °, and it is not related to PLA, but can be caused by the

crystallization of some additive present in the starting material, since, in this work, technical grade PLA was used (there is no data on the exact composition).
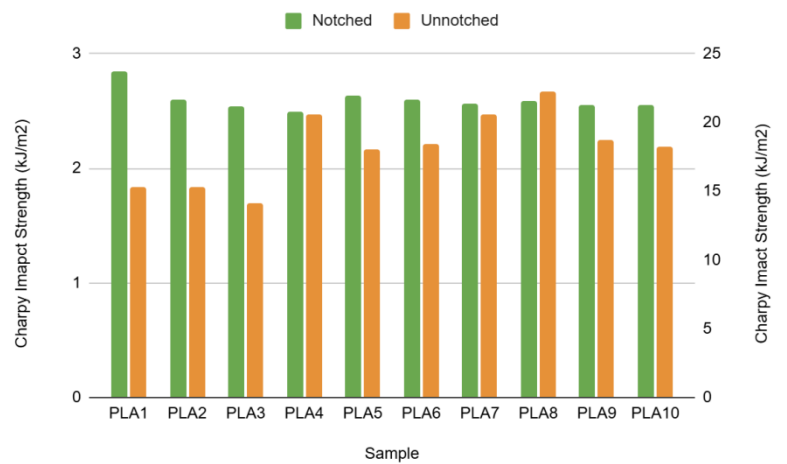

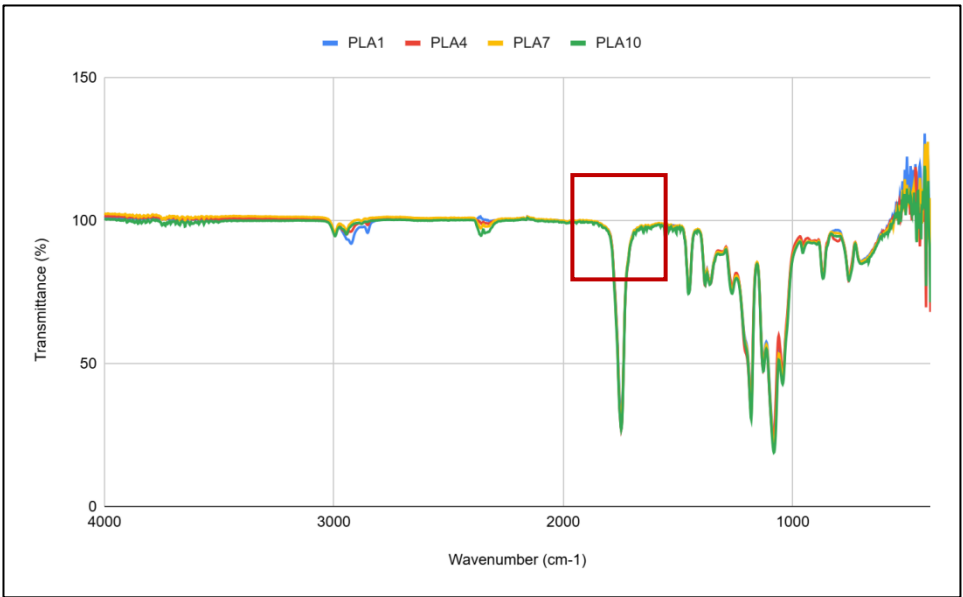**Figure 4.** Impact test results for notched (green) and unnotched (orange) bars.
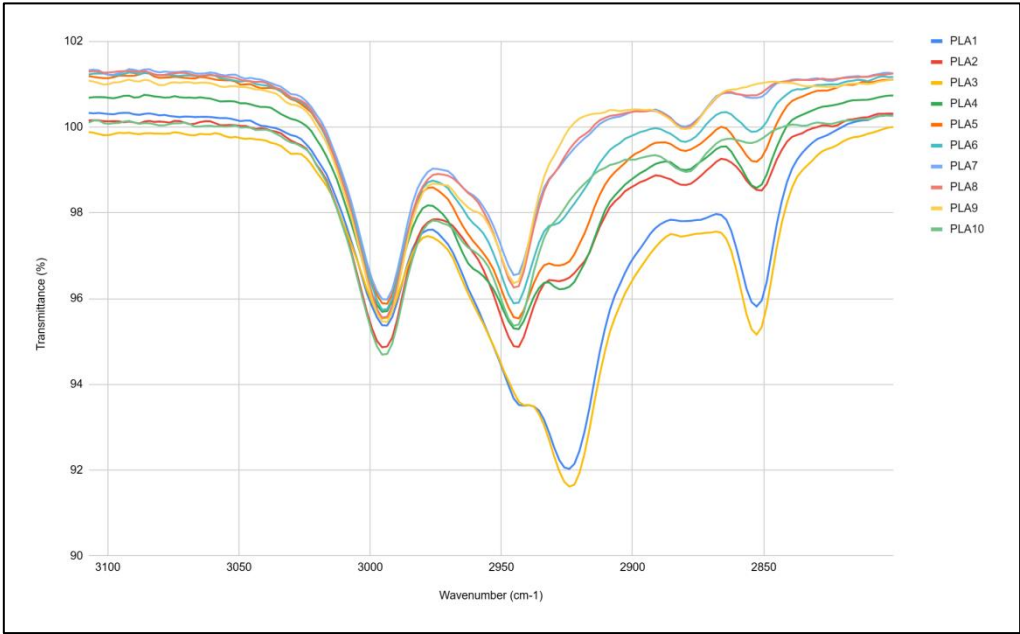

Figure 5. FTIR spectra of selected samples.
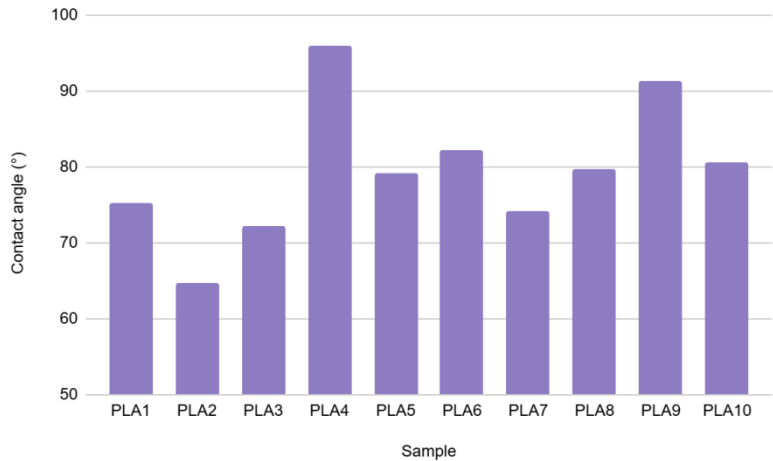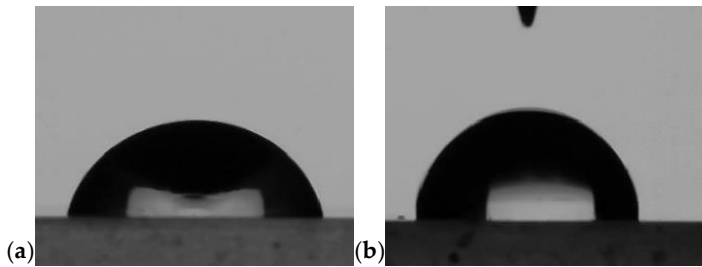
Figure 6. Selection of FTIR peaks.

Figure 7. Contact angle of PLA samples.

The color change development from the first to the tenth injection molding cycle is almost linear. Figure 8. illustrates the increase in ΔE^* values from the second to the tenth sample, which interprets the sample's color difference compared to the reference (in this case, PLA1).
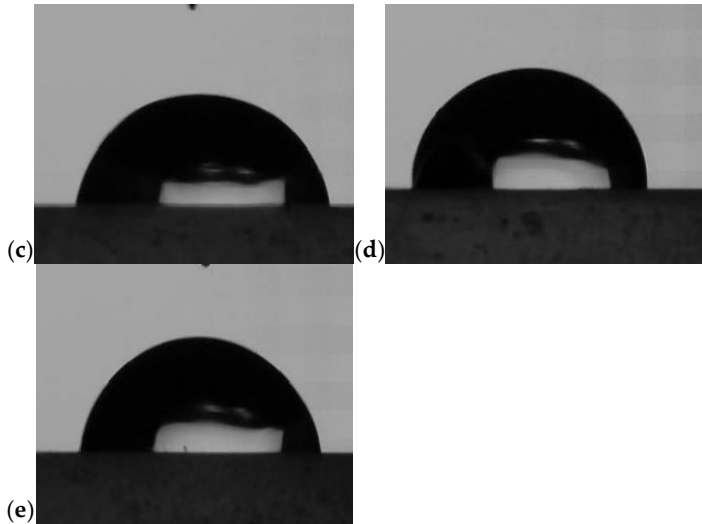

(a)   (b)

Figure 8. Images of water drops on the surface of different samples: a) PLA1, b) PLA4, c) PLA6, d) PLA9 and e) PLA10.

The measured values, along with their respective standard deviations, are presented in Table 5. As the number of recycling cycles increased from PLA1 to PLA10, a clear trend in the optical properties of the samples was observed. The L* parameter consistently decreased from 89.598±0.253 (PLA1) to 84.782±0.232 (PLA10). This reduction in L* indicates that the recycled PLA samples became progressively darker with each successive reprocessing step. Changes were also noted along the chromatic axes. The a* values decreased from −5.878±0.101 (PLA1) to −7.366±0.071 (PLA10), signifying a shift towards a more pronounced green color. Conversely, the b* values showed a consistent increase from 7.98±0.112 (PLA1) to 9.848±0.107 (PLA10), indicating an intensified yellow tone. The combined effect of these changes in a* and b* parameters suggests a distinct yellowish-green discoloration of the PLA material upon repeated recycling.
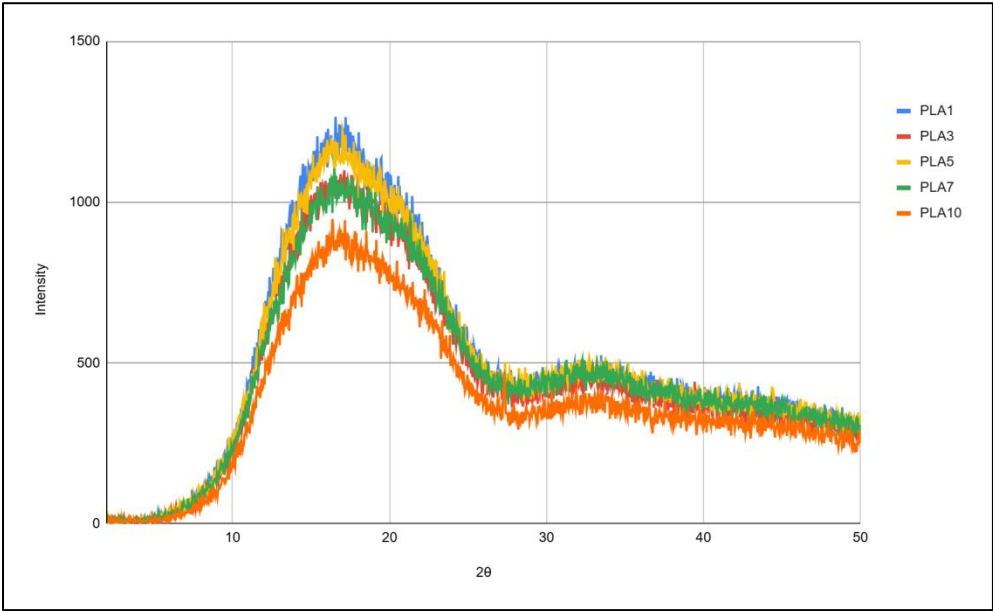

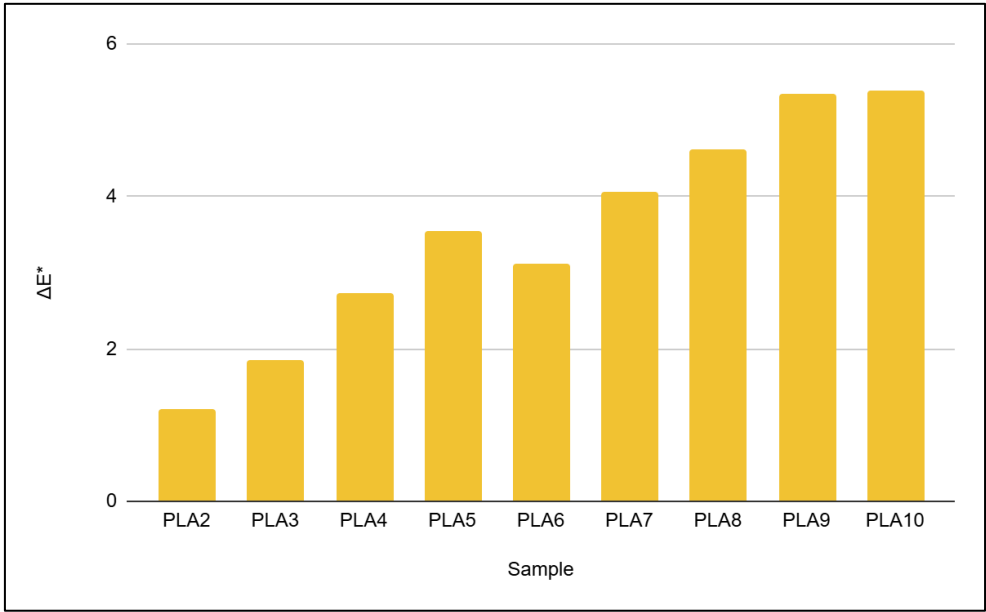**Figure 9.** XRD spectra of PLA samples.

Figure 10. Color change of the samples.

Further analysis of the color saturation, represented by the C* parameter, revealed an increase from 9.912±0.151 (PLA1) to 12.298±0.123 (PLA10). This indicates that the observed color changes are not merely shifts in tone, but also an increase in the vibrancy or intensity of the yellowish-green color. Overall, the colorimetric measurements strongly suggest that the repetitive thermal and mechanical stresses associated with multiple recycling cycles lead to a noticeable degradation of the PLA material, visually manifested as a darkening and a more intense yellowish-green discoloration. These optical changes are likely a direct consequence of polymer degradation mechanisms, such as chain scission and oxidation, which can alter the polymer's light absorption and scattering properties.

Table 5. Colorimetric values of examined samples.

Sample	L*	a*	b*	c*	h*
PLA1	89.598±0.253	-5.878±0.101	7.98±0,112	9.912±0.151	126.382±0.106
PLA2	88.512±0.311	-6.216±0.134	8.43±0.127	10.478±0.181	126.404±0.191
PLA3	87.956±0.499	-6.42±0.185	8.64±0.230	10.762±0.296	126.614±0.169
PLA4	87.166±0.315	-6.67±0.107	8.966±0.156	11.174±0.187	126.658±0.136
PLA5	86.428±0.199	-6.906±0.087	9.216±0.157	11.518±0.166	126.84±0.310
PLA6	86.804±0.132	-6.766±0.067	9.056±0.084	11.308±0.094	126.762±0.270
PLA7	85.982±0.276	-7.016±0.097	9.454±0.107	11.77±0.137	126.572±0.232
PLA8	85.47±0.659	-7.166±0.22	9.596±0.242	11.978±0.324	126.752±0.201
PLA9	84.81±0.688	-7.382±0.238	9.84±0.297	12.3±0.382	126.872±0.065
PLA10	84.782±0.232	-7.366±0.071	9.848±0.107	12.298±0.123	126.796±0.168

5. Conclusions

Within this work, it is shown that multiple mechanical recycling of pre-consumer PLA waste, consisting of 10 cycles of injection molding and milling, does not significantly change the properties of the resulting PLA compared to the starting material without the use of additives or virgin material. An increase in MFI, which indicates a decrease in molecular weight, does not significantly key mechanical properties such as tensile strength, flexural strength, and modulus, as well as thermal properties like Tg, Tm, and degradation temperature, remained stable across cycles. This suggests that multiple mechanical reprocessings do not significantly impair the material's functional performance, especially for non-critical applications. The main drawback of recycling is the color

change that develops from the first to the tenth cycle, making the material more yellow and dull. This color change, driven by polymer degradation mechanisms such as chain scission and oxidation, may limit the reuse of material in aesthetically sensitive applications unless color stabilization strategies are introduced. Overall, these findings reinforce the feasibility of closed-loop mechanical recycling of PLA industrial waste for sustainable material management, providing valuable insights for advancing circular economy practices in the processing of bioplastics.

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Institutional Review Board Statement: Not relevant.

Data Availability Statement: The Dataset is available on request from the authors (alexm@uns.ac.rs, rebeka.lorber@ftpo.eu).

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

Abbreviations

The following abbreviations are used in this manuscript:

PLA	Poly(lactide)
EoL	End-of-Life
MFI	Melt flow index
DSC	Differential scanning calorimetry
Tc	Crystallization temperature
ΔHc	Crystallization enthalpy
Tm	Melting temperature
ΔHm	Melting enthalpy
Xc	Relative crystallinity
ΔHf	Metling enthalpy of 100% crystalline PLA
TGA	Thermo-gravimetric analysis
DMA	Dynamic-mechanical analysis
FTIR	Infrared spectroscopy with Fourrier transformation
XRD	X-ray diffraction
Td_1	Degradation temperature
Tg	Glass transition temperature

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