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Rheological and mechanical properties of recycled polyethylene films contaminated by biopolymer

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Abstract. Nowadays, with the increasing amount of biopolymers used, it can be expected that biodegradable polymers (e. g. PLA, PBAT) may appear in the petrol-based polymer waste stream. However, their impact on the recycling processes is not known yet; moreover, the properties of the products made from contaminated polymer blends are not easily predictable. Therefore, our goal was to investigate the rheological and mechanical properties of synthetic and biopolymer compounds. We made different compounds from regranulates of mixed polyethylene film waste and original polylactic acid (PLA) by extrusion, and injection molded specimens from the compounds. We investigated the rheological properties of the regranulates, and the mechanical properties of the samples. When PLA was added, the viscosity and specific volume of all the blends decreased, and mechanical properties (tensile strength, modulus, and impact strength) changed significantly. Young's modulus increased, while elongation at break and impact strength decreased with the increase of the weight fraction of PLA.

Keywords: recycled polyethylene; polylactic acid; viscosity; pVT diagrams; mechanical properties

1. Introduction

Nowadays most plastic (about 40% of all plastic) is processed by the packaging sector (Gilbert, 2017). The polymers most commonly used for packaging are low density polyethylene (LDPE), linear low density polyethylene (LLDPE), high density polyethylene (HDPE), polypropylene (PP), poly(ethylene terephthalate) (PET), polystyrene (PS) and polyvinyl chloride (PVC) (Gilbert, 2017). The main fields of application of LDPE and LLDPE are reusable bags, trays

1 and containers, agricultural films and food packaging films (Andreoni et al., 2015). According
2 to Plastics News, PET (60%) and PE (35%) accounted for most recycled plastic, due to the
3 short lifetime of packaging materials (Biron, 2017). A wide variety of new products can be
4 produced from plastic waste, therefore it has to be collected and recycled (Andreoni et al., 2015,
5 Sharma and Bansal, 2016).

6 However, lately there has been great demand for packaging made from bioplastics. In 2015, the
7 global production of petroleum-based plastics exceeded 300 million tonnes (Emadian et al.,
8 2017). Nevertheless, in 2014, 1.7 million tonnes of bioplastics were manufactured in the world
9 with almost 75% of the volume destined for the packaging market (Biron, 2017; Emadian et
10 al., 2017). According to European Bioplastics, bioplastics have three main groups: biobased,
11 non-biodegradable materials (bio-PE, bio-PET); biobased and biodegradable materials (PLA,
12 starch blends); and fossil-based and biodegradable materials (poly(buthylene-adipate-co-
13 terephthalate) (PBAT)). Shopping bags, mulch films, and yoghurt cans can be made from PLA
14 (Tábi et al., 2010). However, PLA (4.75 \$/kg) is more expensive than PE (1.9 \$/kg) or PET (1.6
15 \$/kg) (Biron, 2017; Emadian et al., 2017).

16 The potential risk of contamination of the collected conventional plastics and the cost of
17 separation have an impact on the quality and processing of recycled materials. For example,
18 PLA and PET cannot be easily or cheaply sorted by sight or by separation methods based on
19 density because their densities are similar (Gent et al., 2009). Therefore, hybrid bioplastics
20 (blends of bio-based and petroleum-based polymers) appeared. While some biopolymers may
21 have little effect on recycled HDPE, the presence of biopolymers can significantly modify the
22 properties of PET (Cornell, 2007; La Mantia et al., 2012). The presence of biopolymers in post-
23 consumer plastic waste makes the recycling of bioplastic blends complicated (Soroudi and
24 Jakubowicz, 2013).

25 Yarahmadi et al., 2016 investigated the effects of reprocessing and recycling on PLA and a
26 PLA/HDPE blend. They used a commercially available (RTP Company) PLA/HDPE blend
27 which contained 39% PLA. To reduce compatibility problems, they used glycidyl methacrylate-
28 grafted PE as compatibilizer. The materials were evaluated after a varying number of processing
29 steps (up to 6 consecutive extrusions). Multiple processing of the blend did not significantly
30 change the elastic modulus, but elongation at break was significantly affected by the number of
31 extrusions. Two basic mechanisms can affect elongation at break. One is the degradation of one
32 or both polymers, which decreases elongation at break (La Mantia, 1996). The second is the
33 increased dispersion of the dispersed phase, which means smaller domain size and lower
34 interfacial tension, which causes an increase in elongation at break (Fortelný et al., 2011). The

1 results of Yarahmadi et al., 2016 indicated that multiple processing of a PLA/HDPE blend
2 caused increased dispersion of the PLA in the HDPE, and thus increased elongation at break.
3 Using SEM, they observed the cryofractured surfaces of the samples and found that the size of
4 the particles and holes in the blend extruded once were in the range of 1.0-1.4 μm , whereas in
5 the blend extruded six times, the range was 0.5-0.9 μm . The DSC thermograms of the blends
6 showed that the components of the blend were not miscible. The blending of PLA with HDPE
7 not only caused the crystallization of the PLA, but also led to a significant decrease in its cold
8 crystallization temperature and a significant increase in its melting peak temperature. HDPE
9 acted as a nucleating agent and lowered the surface free energy barrier toward nucleation and
10 thus initiated crystallization.

11 PLA has several advantages: it is recyclable, compostable and can be produced from renewable
12 resources. In addition, it has good stiffness and strength, but it has some disadvantages too, such
13 as moisture sensitivity, poor impact resistance and low heat deflection temperature. There are
14 numerous methods to modify its properties, such as plasticization, copolymerization, blending,
15 or producing composites (As'habi et al., 2013; Chow et al., 2016; Graupner et al., 2016; Imre
16 et al., 2014; Tábi et al., 2016).

17 Abdolrasouli et al., 2015 studied virgin PLA and PLA/PE blends which contained 10%, 20%
18 and 30% PE. They investigated the effects of PE content on morphology and the linear
19 viscoelastic properties of the melt. The minor phase was a linear low density polyethylene co-
20 polymer (PE) with butane as co-monomer. The samples were prepared with an internal mixer
21 (Brabender Plasticorder W50) at a temperature of 180°C. They studied the linear viscoelastic
22 behavior of the samples in the melt with a rheometric mechanical spectrometer (Paar Physica
23 UDS 200). They performed all the measurements at 180°C in a parallel plate fixture. When the
24 concentration of PE increased from 10% to 20%, the diameter of the dispersed phase increased
25 and the relaxation process became longer, leading to an increase in the storage modulus. The
26 blend containing 30% of PE showed higher elasticity than both PLA and the PE homo-polymer
27 in the low frequencies. Complex viscosity increased with the increase of the weight fraction of
28 PE, even if this effect was not significant in the higher (100-1000 1/s) shear rate range.

29 Only a few publications examine how the presence of biopolymers affects the properties of
30 conventional petrol-based polymers in their blends. However, according to a recent study of
31 Plastics Recyclers Europe, 2017 it is a considerable problem in Southern Europe because there,
32 in mixed film waste, the share of biodegradable plastics (e.g. starch, PLA, polybutylene adipate
33 terephthalate (PBAT)) is higher than in Northern Europe. Film production with recycled
34 plastics from Northern Europe worked normally, but holes and peaks occurred regularly when

1 recycled plastics from Southern Europe were used. The laboratory tests showed that most of
2 the degradation is caused by substances which are used for the production of biodegradable
3 polymers.

4 Nowadays, when mixed polyethylene film waste is recycled, it is a great problem that small
5 amounts (0.5-1.5%) of bioplastics are often mixed into synthetic polymers. The changes in the
6 properties of a product are not easily predictable, but they are very important to know for the
7 manufacturing of products. The novelty of this manuscript, compared to other publications, is
8 that our goal is not to develop a new blend to improve the properties of PLA or PE, but to
9 analyze the influence of bioplastic contamination in the petrol-based polymer waste stream on
10 the recycling process. Therefore, our goal was to investigate the rheological and mechanical
11 properties of synthetic and biopolymer compounds.

12

13 **2. Experimental**

14 **2.1. Materials**

15 LDPE, PP, HDPE and PET are the most common types of plastic processed by the packaging
16 industry and the composition of the plastic fraction of municipal solid waste by polymer type
17 was LDPE (27.9%), HDPE (14%), PP (17.5%) and PET (11.9%) in Europe (Dahlbo et al., 2018;
18 PlasticsEurope, 2016). According to Biron, 2017, PET (60%) and PE (35%) account for most
19 recycled plastic, PP accounting for only 2%. In 2014, PLA was the most used biodegradable
20 bioplastic (Emadian et al., 2017), therefore there is a chance that PE and PLA are mixed in the
21 polymer waste stream.

22 We used regranulates of mixed polyethylene film waste (MPE) (MFR (190°C, 2.16 kg): 0.56
23 g/10 min, density: 0.932 g/cm³), supplied by Jász-Plasztik Ltd. (Hungary). It is a post-industrial
24 mixed film waste, which typically contains 50% HDPE and 50% LDPE. We also used virgin
25 PLA type Ingeo 3100HP (MFR (210°C, 2.16 kg): 24 g/10 min, density: 1.24 g/cm³), supplied
26 by NatureWorks LLC. (USA).

27

28 **2.2. Material preparation and processing**

29 Six different MPE-PLA compounds (regranulates) (100/0, 98/2, 90/10, 75/25, 50/50, 0/100
30 wt/wt%) were produced with a Labtech Scientific LTE 26-48 co-rotating twin-screw extruder.
31 Before extrusion, PLA was dried at 100°C in a Memmert UF1060 hot air drying oven for 90
32 minutes. The screw diameter of the extruder was 26 mm and the length/diameter (L/D) ratio
33 was 48. The temperature profile of the extruder (from intake zone to die) was 190-195-195-

1 200-200-205-210-215-215-220-220-215°C. The melt pressure was 50-55 bar and the
2 throughput was 8 kg/h.

3 Before injection molding, the compounds were dried at 100°C in a Memmert UF1060 hot air
4 drying oven for 3 hours. The injection molded samples were made with an Engel ES 200/45
5 HL-V injection molding machine. Injection rate was 40 cm³/s, holding pressure was 800 bar,
6 holding time was 20 s, residual cooling time was 30 s, and melt and mold temperature were
7 225°C and 25°C, respectively. The mechanical tests were carried out on ISO standard dumbbell
8 shaped tensile specimens.

9

10 **2.3. Methods**

11 Before the rheological tests, the samples were dried at 100°C in a Memmert UNE 200 hot air
12 drying oven for 3 hours. The shear viscosity of the regranulates was measured with an Instron
13 Ceast SR20 capillary rheometer. The testing temperature in the capillary rheometer was set to
14 190°C. We carried out the measurements in the 100-20000 1/s shear rate range, using four
15 different capillaries: L₁ = 5 mm, D₁ = 1 mm; L₂ = 10 mm, D₂ = 1 mm; L₃ = 20 mm, D₃ = 1 mm;
16 L₄ = 30 mm, D₄ = 1 mm. We used Bagley correction to calculate true shear stress and the
17 Rabinowitsch analysis to calculate true shear rate. The specific volume (pVT diagram) of the
18 regranulates was measured with an Instron Ceast SR20 capillary rheometer. We determined the
19 specific volume with the direct method at 6 different pressures (50-1000 bar) and at 22 different
20 temperatures ranging from 230°C to 50°C (Carreau et al., 1997). We tested all the samples at
21 least 8 times; in every sample standard deviation was within 5% (therefore error bars are not
22 visible on the figures).

23 Tensile tests were carried out on an Instron 3369 universal testing machine. We calculated the
24 tensile modulus between 0.05% and 0.25% strain with a crosshead speed of 1 mm/min, and
25 determined tensile strength and elongation at break with a crosshead speed of 10 mm/min. The
26 measurements were performed on ISO 527-2/1B dumbbell shaped specimens with an overall
27 length of 150 mm and a cross-section of 4 mm × 10 mm. In every measurement, we tested at
28 least 5 specimens, and calculated the average value and standard deviation.

29 Impact strength was determined with the Izod impact test on a Zwick 5113.100/01 impact tester,
30 with pendulums of 2.75 J and 22 J. The measurements were performed on 2 mm notched ISO
31 180/A dumbbell shaped specimens with a length of 80 mm and a cross-section of 4 mm × 10
32 mm. The tests were performed at room temperature and at a relative humidity of 50%. In every
33 measurement we tested at least 10 specimens, and calculated the average and standard
34 deviation.

1 After drying the samples, we measured the residual moisture content with an aboni Hydrotracer
2 FLV water content analyzer. The absolute water content of the material was determined with a
3 chemical method, not based on weight loss during heating. The instrument warmed the PLA
4 granules to 140°C, therefore the water evaporated. The gaseous water reacted with the calcium
5 hydride reagent in a cooled area and hydrogen was formed. A gas sensor detected the hydrogen
6 concentration within the reactor and separate sensors detected the moisture of the ambient air,
7 which came into the reactor at the beginning of the measurement. The software considered these
8 in the calculation of the water content of the PLA granules.

9 The morphology of the blends was studied with a light microscope (Zeiss AXIO Imager M2m)
10 and a scanning electron microscope (Zeiss EVO MA 10). The SEM micrographs were taken
11 from cryo-fractured surfaces.

12 The core and the skin layer of the injection molded samples were investigated with a Bruker
13 Tensor 37 Fourier Transform Infrared Spectroscopy (FTIR) equipment (the resolution of the
14 instrument is better than 0.6 cm⁻¹).

16 **2.4. Processing experiments**

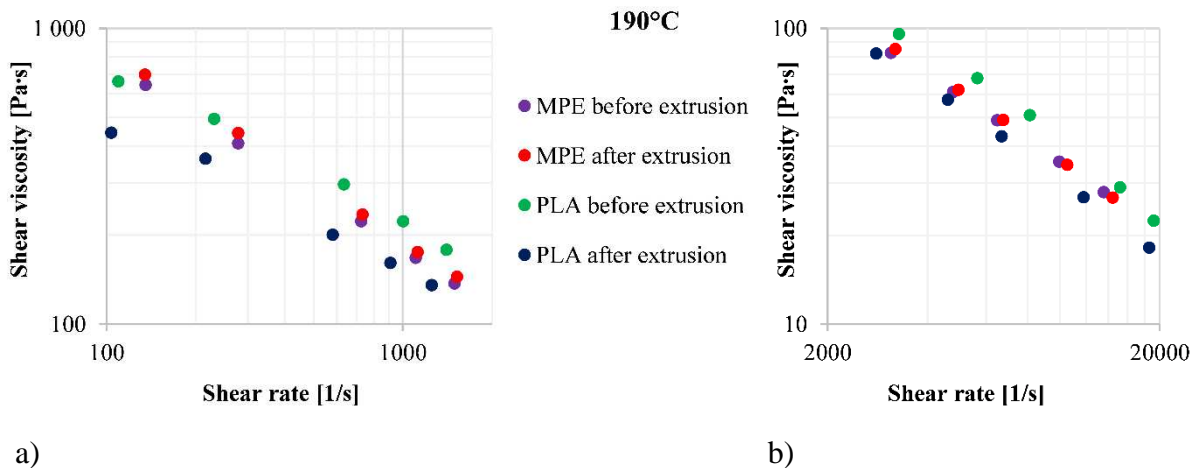
17 In the experiments, MPE was mixed with original PLA (instead of PLA waste), because we did
18 not want to increase the number of influencing factors, we just wanted to discover the effect of
19 biodegradable polymer “contamination”.

20 In the case of PLA, according of the technical data sheet, a moisture content of less than 250
21 ppm (0.025%) is recommended before processing, to prevent hydrolytic degradation. In order
22 to model industrial conditions (PE flakes are usually dried for a short time after the
23 washing/cleaning process), we dried PLA for a shorter time and it had a moisture content of
24 457 ppm before extrusion. Modern film recycling systems (Erema, Starlinger) can process
25 materials (PE, PP) with up to 12% residual moisture. Therefore, drying temperature and/or time
26 can be reduced, and energy consumption, as well as production costs and CO₂ emission can be
27 decreased. However, in the case of hydrophilic biopolymer (PLA) contaminants mixed in
28 synthetic polymers, hydrolytic degradation probably occurs. Melt temperature was 20°C higher
29 than the recommended value for PLA because of the processing melt temperature of MPE,
30 therefore thermal degradation probably occurred. After extrusion the different regranulates
31 looked like typical regranulates.

1 **3. Results and discussion**

2 **3.1. Rheological properties of the regranulates**

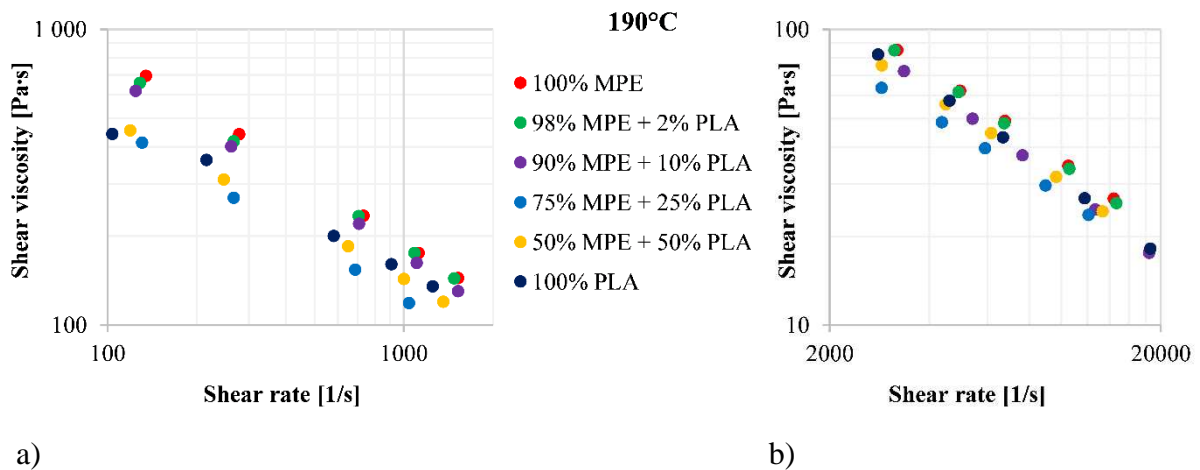
3 Figure 1 shows the shear viscosity curves of MPE and PLA before and after extrusion at 190°C.
4 Extrusion did not affect the viscosity of MPE, but after extrusion, the viscosity of PLA
5 decreased. In the case of PLA, this decrease was due to degradation reactions such as thermal
6 unzipping degradation, oxidative chain scission, inter- or intramolecular transesterification and
7 hydrolysis in the presence of moisture. Residual moisture after the drying process attacks the
8 ester linkages, causing chain scission and a decrease in molecular weight, and as a result,
9 viscosity decreases. Several researchers (Harris and Lee, 2012; Jamshidi et al., 1988;
10 Yarahmadi et al., 2016) came to similar conclusions about the degradation of PLA. Degradation
11 under processing takes place not only in sensitive polymers, such as PET or PLA, but also in
12 polymers that are relatively resistant to degradation, for example PE. In the case of PE, both
13 degradation and cross-linking occur after multiple processing, which can increase melt
14 viscosity. This is also supported by the results of Cornell, 2007.
15



16
17 **Figure 1.** Viscosity curves of MPE and PLA before and after extrusion in the 100-2000 1/s
18 (a) and the 2000-20000 1/s (b) shear rate range

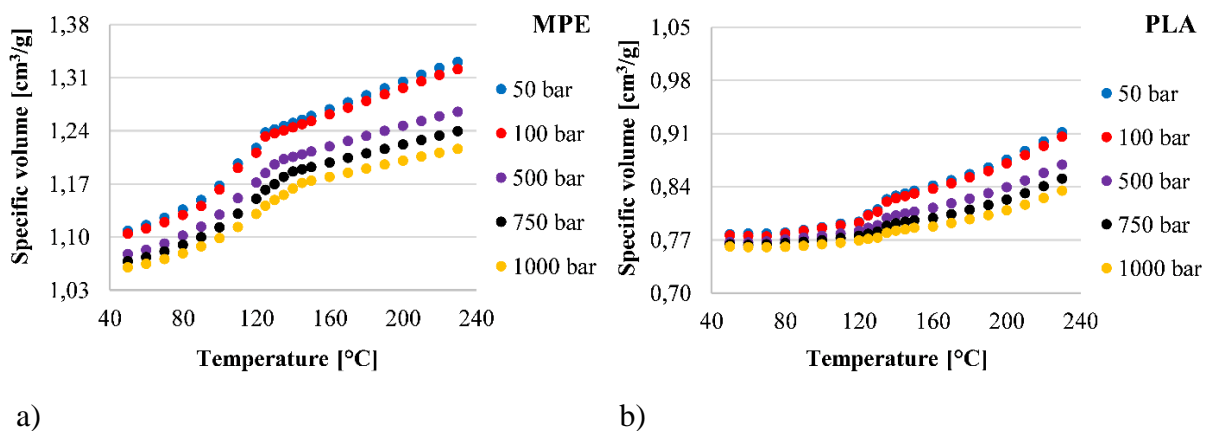
19
20 Figure 2 shows the shear viscosity curves of the six different regranulates in the 100-20000 1/s
21 shear rate range at 190°C. The results indicated that the presence of PLA can significantly affect
22 viscosity. The viscosity curves of all the blends showed a decrease in viscosity when PLA was
23 added but this effect was not significant in the 98/2 blend. The melt viscosities of MPE (100/0)
24 and PLA (0/100) were similar in the higher shear rate range (> 2000 1/s), but the viscosities of
25 the blends did not change according to the linear rule of mixtures. Incompatible blends usually

1 show negative deviation in rheological properties (e.g. viscosity, die swell). Therefore, the two
 2 components were probably incompatible in the melt at least in the concentrations investigated
 3 here. Hamad et al., 2012 found similar results for PLA/LDPE blends in the 0.1-200 1/s shear
 4 rate range. They also observed that the viscosity of the blends indicates negative deviation
 5 according to the linear rule of mixtures.
 6



7 **Figure 2.** Viscosity curves of the different MPE-PLA regranulates in the 100-2000 1/s (a) and
 8 the 2000-20000 1/s (b) shear rate range

9
 10 Figure 3 shows the pvT curves of the extruded MPE and PLA at 5 different pressures. The
 11 specific volume decreased with the increase of pressure. The crystallization of MPE started at
 12 ~125°C, whereas the crystallization of PLA started at ~135°C. Shrinkage (the decrease of
 13 specific volume) was 17% for MPE and 15% for PLA between 230 °C and 50 °C at 50 bar.
 14

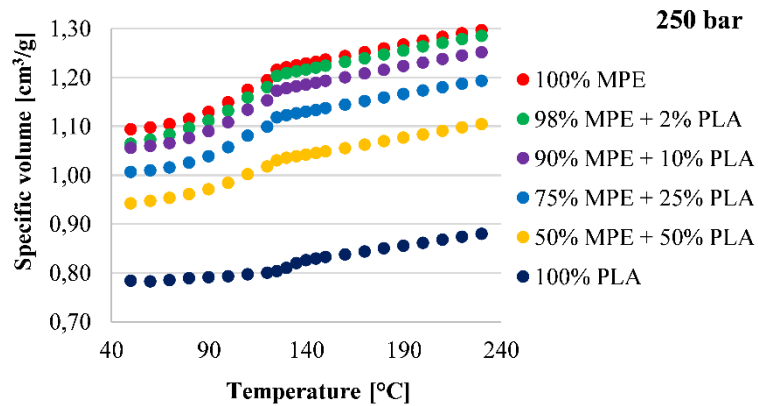


15 **Figure 3.** pvT curves of the extruded MPE (a) and PLA (b) at different pressures

16

1 Figure 4 shows the pvT curves of the different regranulates. The melt pressure for film blowing
 2 is about 200-300 bar in industrial extruders, therefore we analyzed the pvT curves at 250 bar.
 3 Specific volume decreased with the increase of the weight fraction of PLA but this effect was
 4 not significant in the 98/2 blend. This decrease was due to the higher density of PLA. The pvT
 5 curve of blends can be estimated based on the rule of mixtures if the pvT curves of the
 6 components are known.

7



8

9

Figure 4. pvT curves of the different MPE-PLA regranulates at 250 bar

10

11 3.2. Mechanical properties of the injection molded samples

12 Figure 5 shows the tensile stress-strain curves of the six different samples. The curves show
 13 that MPE was flexible and PLA was rigid. The stress-strain curves of all the blends showed a
 14 decrease in elongation, but an increase in tensile strength when PLA was added, but this effect
 15 was not significant in the 98/2 blend.

16

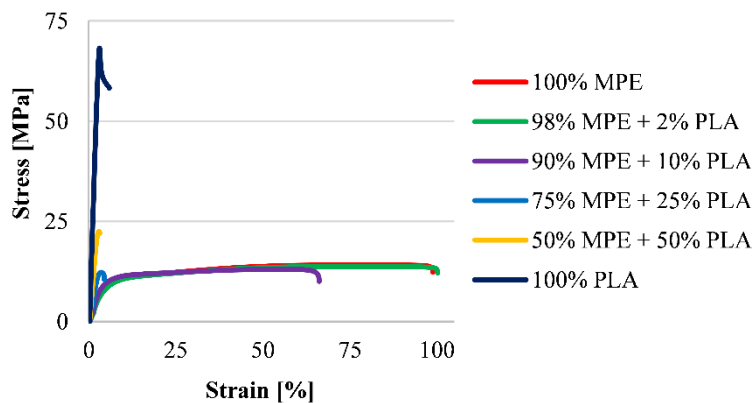
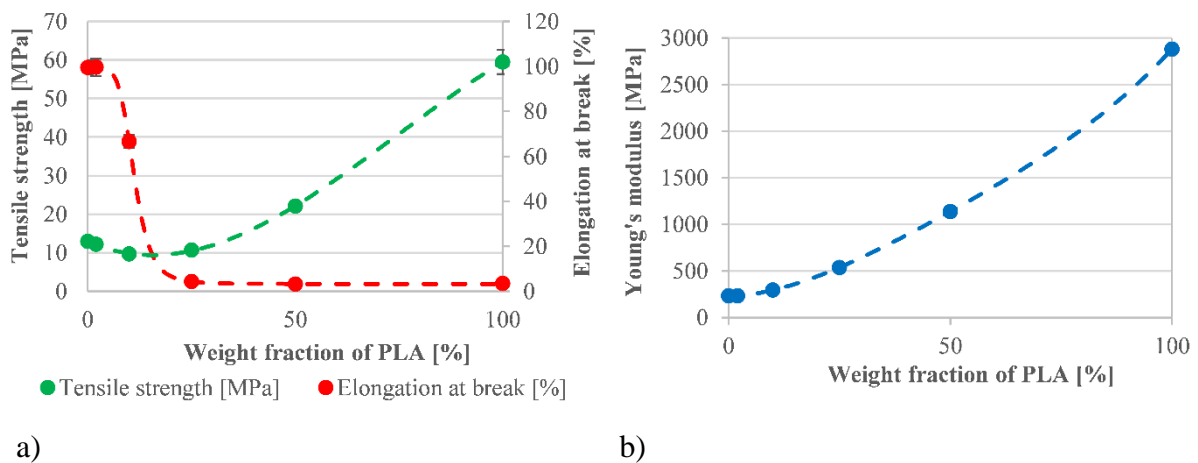


Figure 5. Tensile stress-strain curves of MPE-PLA samples with different weight fractions

18

19 Figure 6 and Table 1 show the mechanical properties of the injection molded samples. The
 20 100/0 and the 98/2 blend had similar mechanical properties. Tensile strength decreased when

1 10 and 25 wt% PLA was added, but doubled when 50 wt% PLA was added. As the weight
 2 fraction of PLA was increased from 0% to 50%, the Young's modulus increased from 232 MPa
 3 to 1137 MPa, while elongation at break decreased from 99.53 % to 3.08 %. In addition to the
 4 presence of a rigid component (PLA), the degradation of PLA and the increased droplet size
 5 (see Figure 9e-h) could also lead to a decrease in elongation at break when the weight fraction
 6 of PLA was increased. The impact strength of the samples greatly decreased when 25 and 50
 7 wt% PLA was added, but the 100/0, 98/2, 90/0 samples were not broken, even when the 22 J
 8 pendulum was used. As the weight fraction of PLA was increased, tensile strength and the
 9 Young's modulus seemed to change along a parabola, while elongation at break decreased
 10 sharply and stayed low, and no tendency could be observed for impact strength. These
 11 mechanical properties of the blends did not change according to the linear rule of mixtures.
 12 Similarly to rheological properties, the polymer blends are typically incompatible if the values
 13 of Young's modulus, elongation at break and stress at break are below the line of linear rule of
 14 mixtures (negative deviation). These results show that PLA and MPE compose an incompatible
 15 blend, which is confirmed by the rheological results. The trend of increasing Young's modulus
 16 and decreasing elongation at break well matched the results of Hamad et al., 2012, who studied
 17 a PLA/LDPE blend.
 18



19 **Figure 6.** Tensile strength and elongation at break (a) and Young's modulus (b) of the
 20 different injection molded MPE-PLA samples
 21
 22
 23

Property	100% MPE	98% MPE + 2% PLA	90% MPE + 10% PLA	75% MPE + 25% PLA	50% MPE + 50% PLA	100% PLA
Impact Strength – Izod [kJ/m ²]	not broken	not broken	not broken	6.07 ± 0.28	2.56 ± 0.11	3.75 ± 0.13

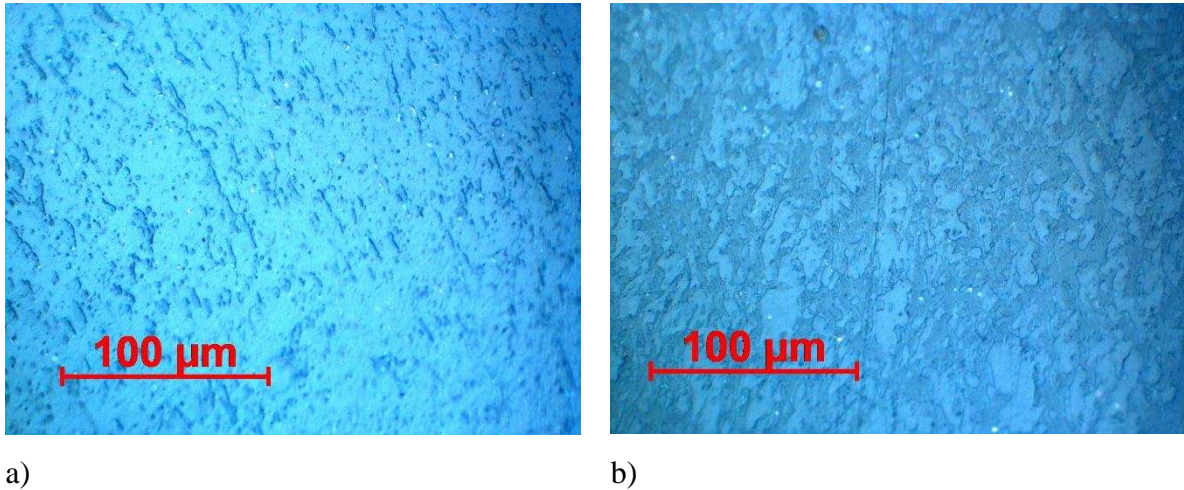
Table 1. Impact strength of the different injection molded MPE-PLA samples

3.3. Structure

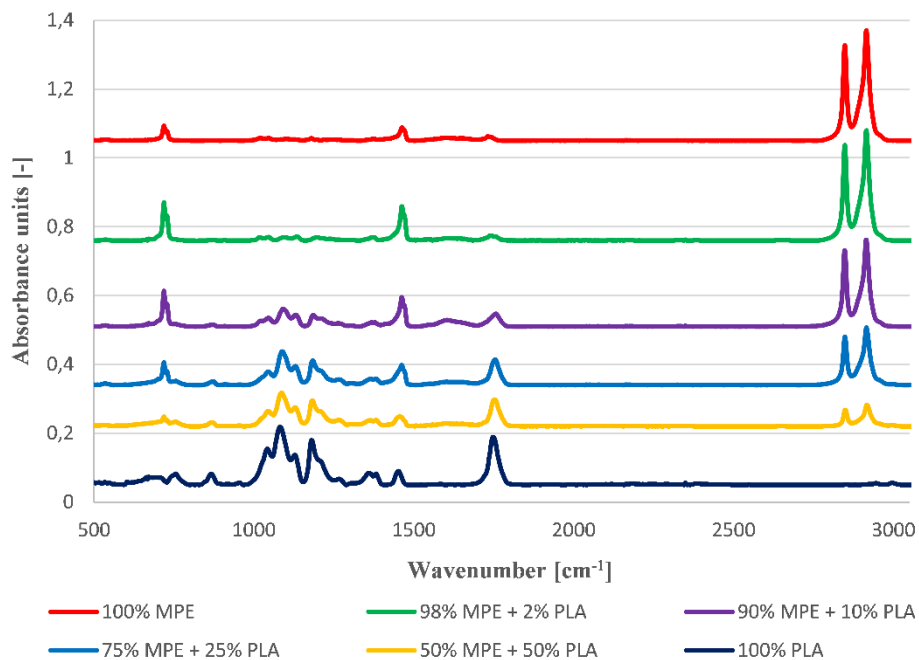
After the tensile tests, it seemed that there was a core part which was flexible and a skin part which behaved more rigidly. To prove this, we analyzed the cross sections of the tensile bars with a light microscope (Figure 7) and an FTIR spectrometer (Figure 8). In the blends which contained 2 and 10 wt% PLA, the dispersed PLA phases were not visible. However, in the case of the 75/25 blend, the dispersed PLA component was visible in the MPE matrix, furthermore, the structure of the 50/50 blend was close to co-continuous. The micrographs were taken from the edge of the cross section, where both components could be found and the components were not clearly layered. The outer surface of the samples was investigated by FTIR. In this analysis the region between 800 and 1800 cm⁻¹ is interesting, where the active ester and methyl groups of PLA are located. A higher intensity in the –C=O ester carbonyl stretch region (~1720-1775 cm⁻¹) was observed with the increase of the weight fraction of PLA. The peak maximum was a wave number of 1752 cm⁻¹; this peak is related to carboxylic end groups formed during the hydrolysis of the ester bonds, which can indicate the degradation of PLA. This is supported by Yarahmadi et al., 2016 and Badia et al., 2012, who observed the influence of multiple processing of neat PLA and PLA blends on degradation. In Figure 8, at the same time, an increase in signal intensity can be observed at the characteristic PLA peak at 1266 cm⁻¹ (–CO bend) and the peak of 1086 cm⁻¹ with the increase of the weight fraction of PLA, which was ascribed to symmetric stretching of the –C–O– band of ester groups. Figure 8 shows that the symmetric deformation of the methyl –CH₃ group occurred at 1381 cm⁻¹ and 1266 cm⁻¹, when PLA content was increased from 10% to 50%. The changes in these peaks were also observed by Ndazi and Karlsson, 2011, when they investigated the hydrolytic degradation of PLA and by Vasile et al., 2017, when they studied PLA bionanocomposites for food packing. In Figure 8, the spectra demonstrated the presence of –CH₃, –CH₂ and C–H groups of highly aliphatic components in the 2800-3000 cm⁻¹ wavenumber range, due to the MPE component. A decrease

1 in signal intensity can be observed at the peak of 2848 cm^{-1} and the peak of 2916 cm^{-1} with the
2 increase of the weight fraction of PLA, which was ascribed to the stretching of the -C-H bond
3 of the alkanes. This is supported by Das and Tiwari, 2018 and Miandad et al., 2017, who
4 observed the pyrolysis of plastic waste.

5



6 **Figure 7.** Changes in the morphology of MPE/PLA blends (a) 25 wt% PLA, b) 50 wt% PLA)
7



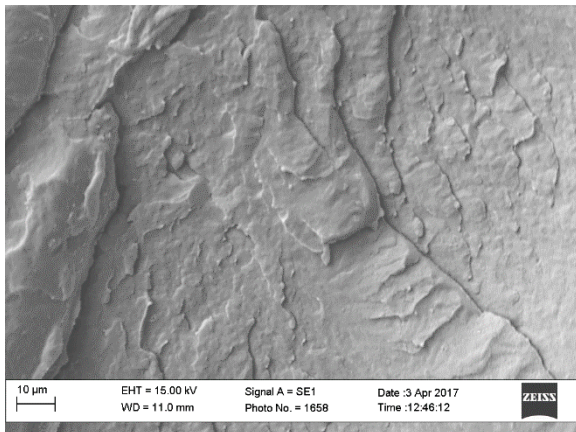
8
9 **Figure 8.** FTIR curves of the different MPE-PLA blends in the $500\text{-}3000\text{ cm}^{-1}$ wavenumber
10 range

11

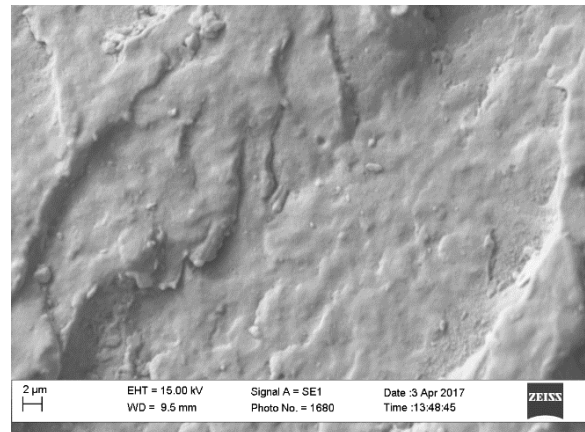
12 We also studied the structure of the blends by SEM (Figure 9). The micrographs were taken
13 from cryo-fractured surfaces. In the case of the 98/2 blend, the dispersed PLA phase formed

1 only a few tenths of a micron droplets (droplet distribution) and then in the 90/10 blend the
2 divided droplets grew a few microns in size. In the blend that contained 25 wt% PLA, the
3 extended dispersed phases were deformed and the spherical droplets were converted to an
4 elongated fibrous shape. Finally, in the 50/50 blend a co-continuous phase formed. These results
5 are consistent with the results of light microscopy. In SEM micrographs, a typical morphology
6 of immiscible polymer blends could be seen if one of the components had a well-defined
7 spherical shape, very broad size distribution and a smooth fracture surface. These observations
8 are also confirmed by rheological and mechanical test results. Kim et al., 2004 had similar
9 results, when they investigated an 80/20 LDPE/PLA blend. Lu et al., 2016 also observed a co-
10 continuous phase morphology in PLA/HDPE blends (60/40 and 50/50) and a typical island-sea
11 type morphology in PLA/HDPE (40/60 and 20/80) blends.

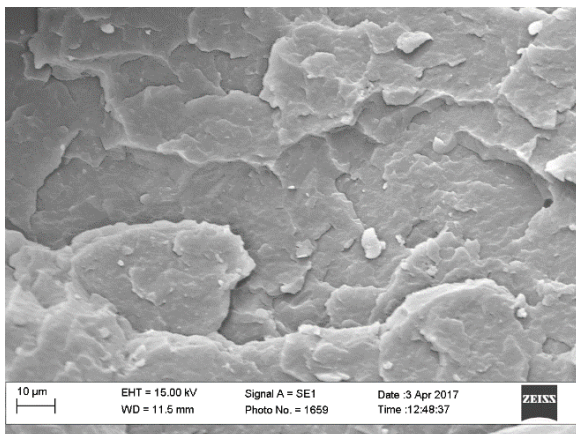
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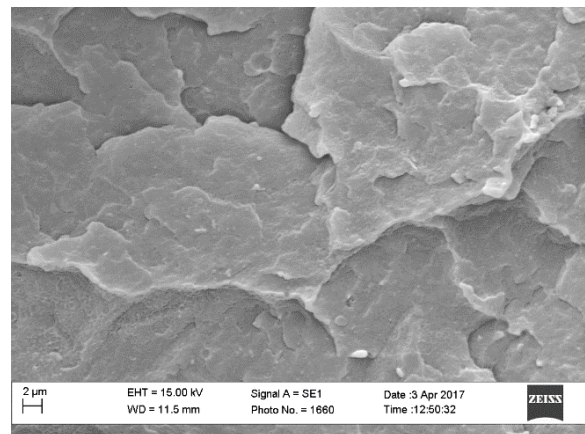
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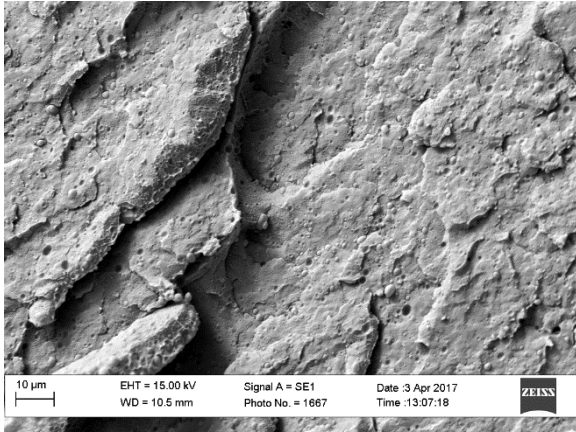
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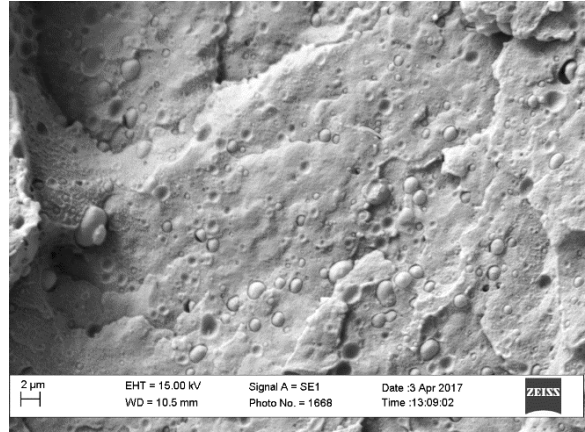
c)



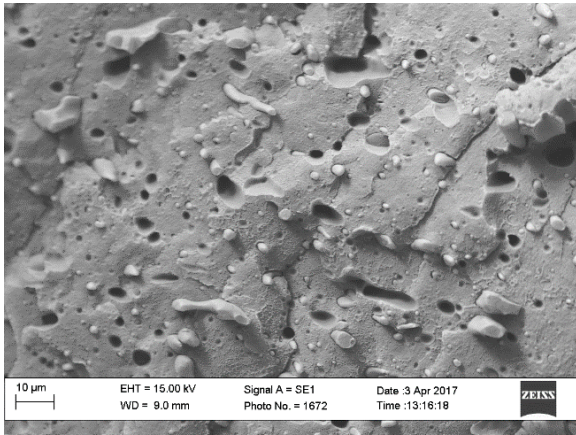
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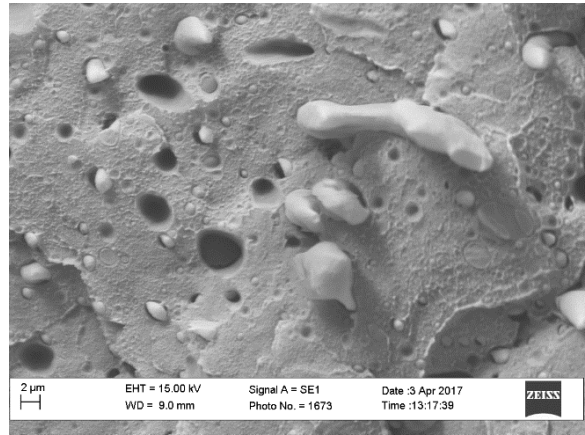
e)



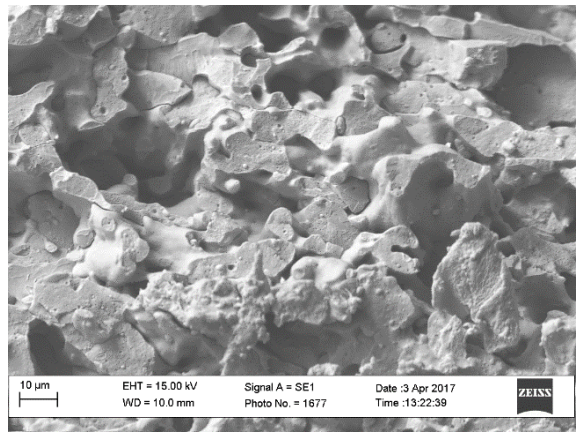
f)



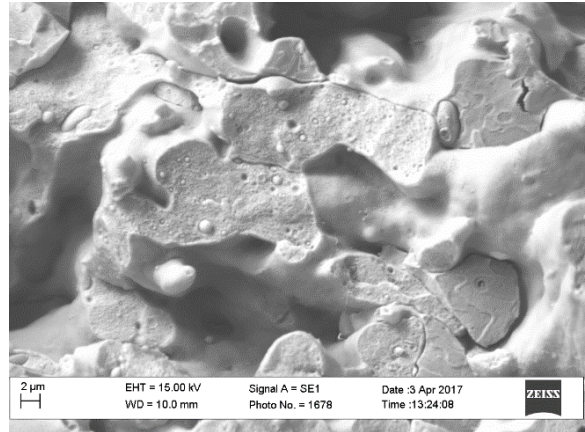
g)



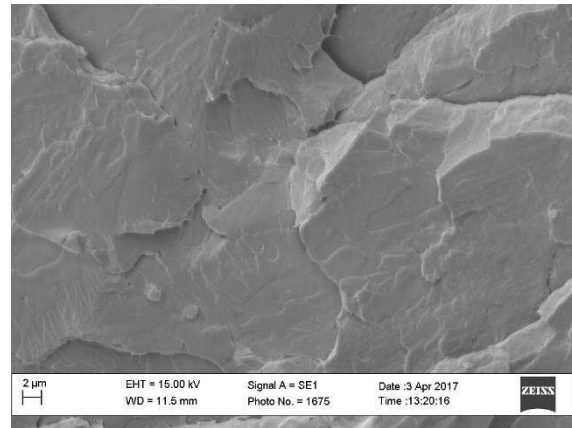
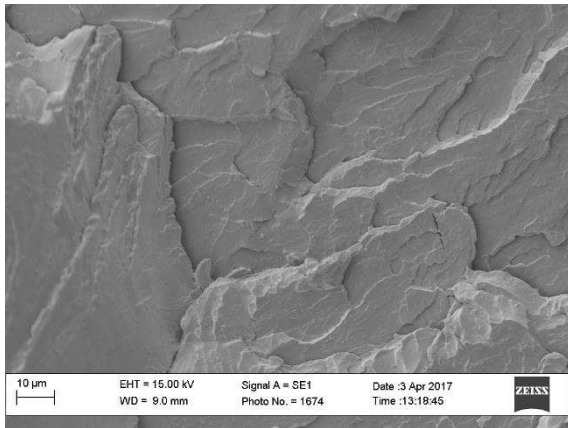
h)



i)



j)



k)

l)

1 **Figure 9.** Changes in the morphology of MPE/PLA blends (a-b) 0 wt% PLA, c-d) 2 wt%
 2 PLA, e-f) 10 wt% PLA, g-h) 25 wt% PLA, i-j) 50 wt% PLA, k-l) 100 wt% PLA)
 3

4

4. Conclusions

5 Nowadays, in the recycling of mixed polyethylene film waste, it is a considerable problem that
 6 small amounts (0.5-1.5%) of bioplastic waste can contaminate synthetic polymer waste.
 7 Developing a method of recycling this post-consumer waste of polymer blends is a major
 8 challenge.

9 We investigated the rheological properties of regranulates and mechanical properties of
 10 injection molded samples.

11 The viscosity curves of all the blends showed a decrease of viscosity when PLA was added.
 12 The melt viscosities of MPE (100/0) and PLA (0/100) were similar, but the viscosities of the
 13 blends did not change according to the linear rule of mixtures. Specific volume decreased with
 14 the increase of the weight fraction of PLA, which is due to the higher density of PLA. The
 15 stress-strain curves showed that MPE was flexible and PLA was rigid. Tensile strength
 16 decreased when 10 and 25 wt% PLA was added, but doubled when 50 wt% PLA was added.
 17 As the weight fraction of PLA was increased from 0% to 50%, the Young's modulus increased
 18 from 232 MPa to 1137 MPa, while elongation at break decreased from 99.53 % to 3.08 %. The
 19 impact strength of the samples greatly decreased when 25 and 50 wt% PLA was added, but the
 20 100/0, 98/2, 90/0 samples were not broken, even when a 22 J pendulum was used. In SEM
 21 micrographs, we observed a co-continuous phase morphology in the 50/50 blend, and a typical
 22 island-sea type morphology in the 98/2, 98/10 and 75/25 MPE/PLA blends. Therefore, the two
 23 components are incompatible, at least in the concentrations investigated.

24 Based on the results, it seems that the blend containing 2% PLA can still be used for film
 25 production, but the other blends are inappropriate for this purpose. The blends that contain more

1 than 2 % of PLA would probably result in a lot of faulty products, due to the immiscibility of
2 the components. These faulty products cannot be recycled, they just go to a landfill or are used
3 for energy production. The miscibility of the blends can probably be improved by adding
4 compatibilizers, therefore products with a simple geometry and low mechanical loads (e. g.
5 flowerpot, speed bump etc.) can be produced from these contaminated blends. The applicability
6 and cost-effectiveness of compatibilizers needs further research. Our opinion is that
7 biodegradable plastics should be collected into a separate waste stream and then recycled (if
8 possible) or composted.

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