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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.

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

However, lately there has been great demand for packaging made from bioplastics. In 2015, the global production of petroleum-based plastics exceeded 300 million tonnes (Emadian et al., 2017). Nevertheless, in 2014, 1.7 million tonnes of bioplastics were manufactured in the world with almost 75% of the volume

destined for the packaging market (Biron, 2017; Emadian et al., 2017). According to European Bioplastics, bioplastics have three main groups: biobased, non-biodegradable materials (bio-PE, bio-PET); biobased and biodegradable materials (PLA, starch blends); and fossil-based and biodegradable materials (poly(buthylene-adi pate-co-terephthalate) (PBAT)). Shopping bags, mulch films, and yoghurt cans can be made from PLA (Tábi et al., 2010). However, PLA (4.75 \$/kg) is more expensive than PE (1.9 \$/kg) or PET (1.6 \$/kg) (Biron, 2017; Emadian et al., 2017).

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

Yarahmadi et al. (2016) investigated the effects of reprocessing and recycling on PLA and a PLA/HDPE blend. They used a commercially available (RTP Company) PLA/HDPE blend which contained 39% PLA. To reduce compatibility problems, they used glycidyl

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methacrylate-grafted PE as compatibilizer. The materials were evaluated after a varying number of processing steps (up to 6 consecutive extrusions). Multiple processing of the blend did not significantly change the elastic modulus, but elongation at break was significantly affected by the number of extrusions. Two basic mechanisms can affect elongation at break. One is the degradation of one or both polymers, which decreases elongation at break (La Mantia, 1996). The second is the increased dispersion of the dispersed phase, which means smaller domain size and lower interfacial tension, which causes an increase in elongation at break (Fortelný et al., 2011). The results of Yarahmadi et al. (2016) indicated that multiple processing of a PLA/HDPE blend caused increased dispersion of the PLA in the HDPE, and thus increased elongation at break. Using SEM, they observed the cryofractured surfaces of the samples and found that the size of the particles and holes in the blend extruded once were in the range of 1.0–1.4  $\mu\text{m}$ , whereas in the blend extruded six times, the range was 0.5–0.9  $\mu\text{m}$ . The DSC thermograms of the blends showed that the components of the blend were not miscible. The blending of PLA with HDPE not only caused the crystallization of the PLA, but also led to a significant decrease in its cold crystallization temperature and a significant increase in its melting peak temperature. HDPE acted as a nucleating agent and lowered the surface free energy barrier toward nucleation and thus initiated crystallization.

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

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

Only a few publications examine how the presence of biopolymers affects the properties of conventional petrol-based polymers in their blends. However, according to a recent study of *Plastics Recyclers Europe* (2017) it is a considerable problem in Southern Europe because there, in mixed film waste, the share of biodegradable plastics (e.g. starch, PLA, polybutylene adipate terephthalate (PBAT)) is higher than in Northern Europe. Film production with recycled plastics from Northern Europe worked normally, but holes and peaks occurred regularly when recycled plastics from Southern Europe were used. The laboratory tests showed that most of the degradation is caused by substances which are used for the production of biodegradable polymers.

Nowadays, when mixed polyethylene film waste is recycled, it is a great problem that small amounts (0.5–1.5%) of bioplastics are often mixed into synthetic polymers. The changes in the properties of a product are not easily predictable, but they are very

important to know for the manufacturing of products. The novelty of this manuscript, compared to other publications, is that our goal is not to develop a new blend to improve the properties of PLA or PE, but to analyze the influence of bioplastic contamination in the petrol-based polymer waste stream on the recycling process. Therefore, our goal was to investigate the rheological and mechanical properties of synthetic and biopolymer compounds.

## 2. Experimental

### 2.1. Materials

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

We used regranulates of mixed polyethylene film waste (MPE) (MFR (190 °C, 2.16 kg): 0.56 g/10 min, density: 0.932 g/cm<sup>3</sup>), supplied by Jász-Plasztik Ltd. (Hungary). It is a post-industrial mixed film waste, which typically contains 50% HDPE and 50% LDPE. We also used virgin PLA type Ingeo 3100HP (MFR (210 °C, 2.16 kg): 24 g/10 min, density: 1.24 g/cm<sup>3</sup>), supplied by NatureWorks LLC. (USA).

### 2.2. Material preparation and processing

Six different MPE-PLA compounds (regranulates) (100/0, 98/2, 90/10, 75/25, 50/50, 0/100 wt/wt%) were produced with a Labtech Scientific LTE 26–48 co-rotating twin-screw extruder. Before extrusion, PLA was dried at 100 °C in a Memmert UF1060 hot air drying oven for 90 min. The screw diameter of the extruder was 26 mm and the length/diameter (L/D) ratio was 48. The temperature profile of the extruder (from intake zone to die) was 190–195–195–200–200–205–210–215–215–220–220–215 °C. The melt pressure was 50–55 bar and the throughput was 8 kg/h.

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

### 2.3. Methods

Before the rheological tests, the samples were dried at 100 °C in a Memmert UNE 200 hot air drying oven for 3 h. The shear viscosity of the regranulates was measured with an Instron Ceast SR20 capillary rheometer. The testing temperature in the capillary rheometer was set to 190 °C. We carried out the measurements in the 100–20,000 1/s shear rate range, using four different capillaries: L<sub>1</sub> = 5 mm, D<sub>1</sub> = 1 mm; L<sub>2</sub> = 10 mm, D<sub>2</sub> = 1 mm; L<sub>3</sub> = 20 mm, D<sub>3</sub> = 1 mm; L<sub>4</sub> = 30 mm, D<sub>4</sub> = 1 mm. We used Bagley correction to calculate true shear stress and the Rabinowitsch analysis to calculate true shear rate. The specific volume (pvT diagram) of the regranulates was measured with an Instron Ceast SR20 capillary rheometer. We determined the specific volume with the direct method at 6 different pressures (50–1000 bar) and at 22 different temperatures ranging from 230 °C to 50 °C (Carreau et al., 1997).

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