# Combining polymeric waste streams to improve functional properties of post-consumer mixed polyolefines

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ABSTRACT: Polymeric waste streams can be re-used as secondary material sources for polymer processing by extrusion or injection moulding. One of the major commercially available waste streams is the so called mixed polyolefines (rMPO). These MPO mainly consist of a mixture of different grades of polyethylene and polypropylene. An important practical hurdle for the direct implementation of this waste stream is the inherent immiscibility of polyethylene and polypropylene in the melt, which leads to segregation within the final structure and adversely affects the reproducibility and mechanical properties of the manufactured parts. Melt mixing this primary waste stream with other polymeric waste streams could mitigate these drawbacks. Within the current research, a commercially available rMPO waste stream was characterized in terms of composition, rheological and mechanical properties. This study aims to improve the rheological and mechanical properties of the recycled MPO waste stream by melt blending with a polyolefin elastomer (POE) and three different post-industrial waste streams. The added waste streams are high quality post-industrial waste streams i.e. recycled PA/PE/PA foils, PET-fibres and PET-PE packaging trays. This blending only caused a slight improvement in mechanical properties. Different results were obtained depending on the type of waste stream used. The melt flow rate (MFR) on the other hand, was increased to the same level as a chosen PP target with only 2.5 wt% of POE.

## 1 INTRODUCTION

Plastics are one of the most commonly used materials and therefore, they have become a significant part of our lives. Polymers are used in a wide variety of applications, from packaging material to automotive parts and other high-tech applications. Polyolefins, and mainly low-density polyethylene (LDPE), high-density polyethylene (HDPE) and polypropylene (PP) are together responsible for a huge share of the European demand for plastics. PE accounts for around 30% of all plastic demand. PP is good for another 19%. The increasing demand also results in significant waste streams (PlasticsEurope 2016).

Mechanical recycling is one of the most widely practiced recycling method. Generally this process includes washing, sorting, remelting and reprocessing the waste into a new final product. During the sorting process, the different plastic materials are divided from one another. This is a crucial step in the recycling process because most polymers are immiscible. Blends of immiscible materials undergo phase separation which ultimately affect the mechanical properties. Examples of sorting processes around Europe were recently reviewed by Ragaert et al. (Ragaert, Delva et al. 2017). Despite the importance of an accurate separation, a full separation of plastic waste into the individual components is economically and technically not feasible and sometimes quite impossible. This is often the problem with polyolefins. Due to their similar structure and little difference in density, HDPE, LDPE and PP cannot be separated using the existing sorting technologies. These polymers are therefore reprocessed as a blend and are referred to as the so-called 'hard' mixed polyolefins (MPO). (Ragaert, Delva et al. 2017).

Despite the fact that the polyethylenes and polypropylenes in these MPO's are quite similar (i.e. they all consist of carbon and hydrogen atoms), even these mixtures are often not miscible due to differences in molecular structure and form heterogeneous blends which in turn have quite low properties due to the formation of weak interfaces compared to virgin polymers (Hubo, Leite et al. 2014). Therefore, these MPO waste streams are nowadays mechanically recycled mainly into 'low-quality' products such as garden furniture, outdoor flooring and traffic signalization elements by industry (Karlsson 2004).

To extend the application field of recycled polyolefins, researchers have been looking for various methods to upcycle the waste streams. The most commonly used method is melt blending. It refers to blending recycled plastics with similar virgin or different types of recycled plastics in the melt process (Delva, Cardon et al. 2018). Furthermore, adding an additive like a compatibilizer can also be possible. (Ragaert, Hubo et al. 2017).

The main goal of this study is to upcycle a recycled MPO waste stream. In a first part, the rheological properties are improved by adding a polyolefin elastomer (POE). Therefore different percentages of POE are added to the rMPO matrix. The rheology is evaluated through basic melt flow rate (MFR) measurements.

In the second part of this research, three different waste streams are melt blended with the rMPO to improve the mechanical properties. The added waste streams are high quality post-industrial waste streams i.e. recycled PA/PE/PA foils, PET-fibres and PET-PE packaging trays.

## 2 MATERIALS AND METHODS

### 2.1 *Materials*

The post-consumer recycled mixed polyolefins (r-MPO) were delivered by Govaerts Recycling nv. The composition was determined by using Fourier Transform Infrared Spectroscopy (FTIR) and can be found in Figure 1. The dominant polymeric fractions are HDPE and PP.

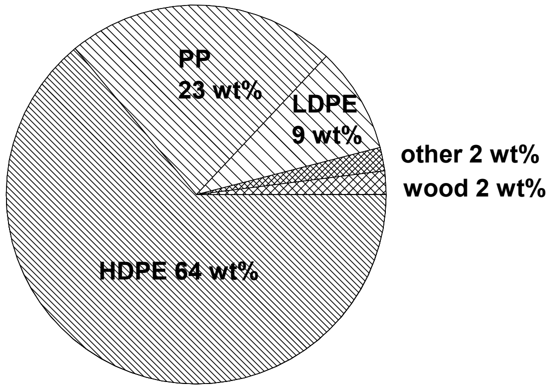


Figure 1. Pie chart of the composition of the rMPO determined by FTIR analysis.

The other recycled materials used are PA/PE/PA recycled foils (Segers & Balcaen), a PET-fibre mix (Belrey Fibres nv) and recycled PET-PE trays with topfoils (Ter Beke). All of them are post-industrial waste streams. The PA/PE/PA (i.e. PA6 and LLPDE) recycled foils are multilayer films used as a film barrier for the production of sheet moulding compounds. The ratio between the different polymers is approximately 50/50 vol% PA/PE. Between the different layers there are tie layers present, containing an adhesive. The foils were reprocessed and delivered in pellet form. The recycled PET-fibres were provided by Belrey Fibres nv and had an average length of 5 mm. The PET-mix is a mixture of recycled PET-streams from unknown origin.

As virgin material, a PP based elastomer (POE) (Vistamaxx 8880, ExxonMobil) with very low viscosity (1200 mPa.s, at 190°C) was used to improve the flow of the rMPO.

### 2.2 *Sample preparation*

In the first part of this research, different POE percentages (2.5 wt%, 5 wt%, 7.5 wt%, 10 wt%) were added to the rMPO matrix and melt blended using a co-rotating twin-screw extruder ZSK18 from Coperion. The temperature profile was set at 180, 210, 210, 215, 215, 220, 220, 230, 230°C, the screw speed was maintained at 150 rpm. After extrusion, the filament was cooled down to room temperature and pelletized.

For the second part of this study, the rMPO and the different post-industrial waste streams were used to prepare blends with a weight ratio of 80/20. After drying all post-industrial waste streams for 24 h at 60°C, they were manually mixed with the rMPO. These mixtures were then melt blended using the same twin-screw extruder and temperature profile as in the first part of this research, except for the rMPO/PET-mix blend where a different temperature profile up to 260°C was used to allow extrusion. After extrusion, all blends were cooled down and granulated.

Finally, the different blends were injection moulded into test bars using an Engel 80T. The temperature profile was set at 200, 210, 220, 230°C. All test samples were then left to condition at room temperature (23 ± 1°C and relative humidity of 50 ± 10%) for at least two days prior to testing.

### 2.3 *Characterization*

To have an indication of the rheology of the rMPO/POE blends, the melt flow rate (MFR, g/10 min) was measured according to ISO 1133 at a temperature of 250°C and a load of 2.16 kg. The reported values are an average of at least five measurements.

Tensile properties were obtained using an Instron 5565 dynamometer according to ISO 527. A clip-on extensometer was used to precisely measure the strain for the determination of the Young’s modulus with a crosshead speed of 2 mm/min. The speed was raised to 10 mm/min from 4% strain to sample breakage.

The Charpy impact properties were determined using a Tinius Olsen IT503 with a pendulum of 5 J. The samples were notched 2 mm and tested according to ISO 179.

All mechanical tests were performed at room temperature. Minimum five specimens were tested for each sample and the average values with standard deviations are reported.

## 3 RESULTS

### 3.1 *Improving the melt flow with POE elastomer*

The MFR values and their respective standard deviations are plotted in Figure 2. The MFR value of a chosen target value is also plotted. This target value matches the value of a PP with an optimal MFR for the foreseen application

In general, adding the POE increases the MFR. The results also show an increasing trend with increasing amount of POE. By adding 7.5 wt%, the MFR of the rMPO is already doubled. This increase originates from the very low viscosity of the POE. It is also clear that the addition of 2.5 wt% of POE is already sufficient to increase the MFR of the rMPO to the same level as the chosen PP target.

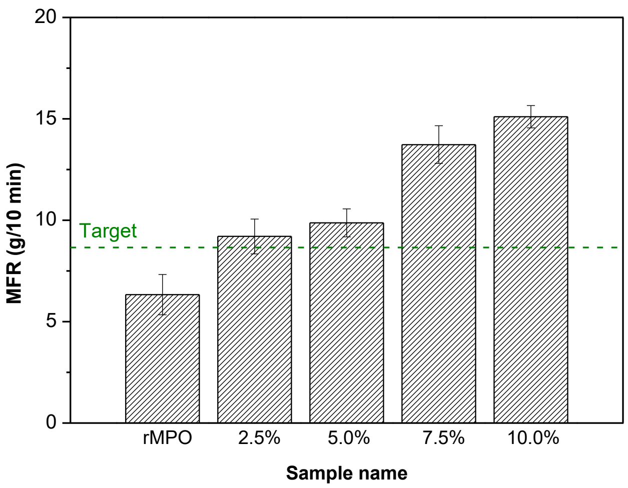


Figure 2. MFR of rMPO plotted next to rMPO blends with different POE weight percentages. Green dashed line represents the target value (8.65 ± 0.52 g/10 min). Error bars represent one standard deviation.

### 3.2 *Improving the mechanical properties with other recycled waste streams*

The results of the mechanical characterization of the rMPO, the individual post-industrial waste streams and the corresponding blends are given in Table 1. Only the values for the PET-mix are missing. It was not possible to obtain the mechanical properties of the PET-mix as such.

#### 3.2.1 *PA/PE/PA recycled foils*

The PA/PE/PA recycled foils have better mechanical properties compared to the rMPO as can be seen in Table 1. The high impact strength (90.9 ± 9.4 kJ/m²) is remarkable. This could first of all be attributed to the presence of the PA. But it can also be due to the presence of various adhesives in the tie layers between the different layers of the foils which could help improve adhesion even after recycling. Therefore it is possible that not only PA but also PE contributes to the impact strength.

Blending rMPO with the recycled PA/PE/PA foils results in improved tensile properties. Tensile strength and strain at break values are increased when compared to the rMPO. Only the stiffness does not show any improvement. In accordance to the strain at break, the impact strength also increased.

Table 1: Mechanical properties for the blended materials.

| | $E_t$ ± stdev [MPa] | $\sigma_t$ ± stdev [MPa] | $\varepsilon_b$ ± stdev [%] | Impact strength ± stdev [kJ/m²] |
|---|---|---|---|---|
| rMPO | 750 ± 31 | 15 ± 1 | 57 ± 35 | 13.4 ± 1.8 |
| PA/PE/PA | 957 ± 102 | 35 ± 1 | 384 ± 18 | 90.9 ± 9.4 |
| +20% PA/PE/PA | 708 ± 127 | 18 ± 1 | 270 ± 160 | 25.3 ± 2.9 |
| +20% PET-mix | 1036 ± 95 | 15 ± 1 | 9 ± 2 | 4.8 ± 0.5 |
| PET-PE | 1677 ± 91 | 31 ± 1 | 14 ± 10 | 4.0 ± 0.3 |
| +20% PET-PE | 1016 ± 77 | 15 ± 1 | 21 ± 7 | 8.1 ± 0.2 |

#### 3.2.2 *Recycled PET-mix*

The rMPO/PET-mix blend has a greater stiffness compared to the rMPO. Although PET generally has a higher strength than PE and PP, no increase in tensile strength is observed for the blend. A possible explanation can be found in the phenomenon of cavitation. Especially in the case of the rMPO/PET-mix blend, it is possible that during tensile drawing at low strain deformations, both materials will contribute to the elastic deformation. But when plastic deformation of the rMPO takes place, the matrix can pull off of the PET spheres due to cavitation and decohesion. PET will not undergo plastic deformation and therefore, will not contribute to the strength of the blend.

Furthermore, the strain at break and impact strength are negatively affected by the presence of PET. The loss in toughness is a result of the brittle nature of PET.

#### 3.2.3 *Recycled PET/PE packaging trays and foils*

The same trends are observed when looking at the rMPO/PET-PE blend. The tensile modulus is improved. But the tensile strength did not change. A possible explanation can be the occurrence of cavitation, as mentioned before. Furthermore, the blend becomes more brittle. This can be seen in the lowering of the strain at break and impact strength. Adding PET can lower the chain flexibility of the PE and PP chains present in the matrix, which can lead to a possible reduction in toughness (Razavi, Shojaei et al. 2011).

#### 3.2.4 *Comparison between different blends*

Blending of different materials often results in a trade-off relationship between two important parameters i.e. stiffness and impact strength. This important relationship is therefore plotted in Figure 3.

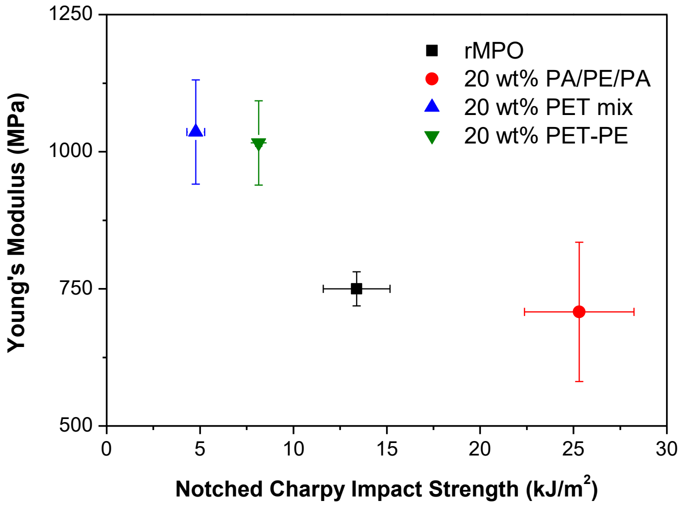


Figure 3. Plot of Young's modulus versus Notched Impact Strength. Error bars represent one standard deviation.

It is clear that adding PET containing waste streams (PET mix and PET-PE) increases the Young's modulus of the rMPO, while reducing the impact strength. On the other hand, adding the PE/PA multilayered foil does the opposite i.e. increasing the impact strength and reducing the Young's modulus.

## 4 CONCLUSION

This study investigates the possibility to improve the rheological and mechanical properties of a recycled MPO waste stream by melt blending with a POE and different post-industrial waste streams.

Adding different weight percentages of POE to the rMPO matrix showed an increase in MFR. There was also an increasing trend observed when the amount of POE increased. The MFR results showed that 2.5 wt% was already sufficient to improve the flow to the same level as a chosen PP target.

In the second part of this research the mechanical properties of different blends containing rMPO and 20 wt% of different post-industrial waste fractions were investigated. Adding PA/PE/PA recycled foils improved the tensile properties, except the stiffness. Furthermore, the impact strength was raised and even almost doubled. The blends which contained a PET fraction, added through the PET-mix or the PET-PE packaging trays, only showed an improvement in stiffness. Strain at break and impact strength were reduced.

Overall it can be concluded that it is possible to improve the properties of rMPO. Although, improving the mechanical properties is only possible to a certain extent depending on the starting properties of the recycled waste stream.

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