

# Remarkable role of experimental olefin-maleic-anhydride copolymer based compatibilizing additives in PET bottle recycling

Nagy B.<sup>1,\*</sup>, Varga Cs.<sup>1</sup>

<sup>1</sup>Institutional Department of MOL Hydrocarbon and Coal Processing, University of Pannonia, Veszprém 8200, Hungary

\*corresponding author: e-mail:nagy.bianka@mk.uni-pannon.hu

## Abstract

Over the past 50 years demand for plastics drastically increased worldwide resulting in plastic wastes causing serious environmental problems. The main market sector of European plastics industry is the packaging industry most of which is polyolefins and poly(ethylene-terephthalate). Polymer blends based on waste resources can solve the issues of recycling. We have studied rheological and tensile properties of three types of PET/engineering thermoplastic blends (PET/PC, PET/PA and PET/ABS) produced with different processing techniques. Miscibility of components of blends is limited leading to weak mechanical properties such as low tensile strength and/or elongation at break. Due to that phenomenon compatibilizing additives are also required. As compatibilizing additives olefin-maleic-anhydride copolymer based additives have been used in our experiments. Structure of additives differed from each other both in ratio and length of carbon chains of compounds linked to maleic-anhydride groups. Blends have been studied with PET content ranging from 10 % to 90 %. As an outstanding result we have managed to achieve improving mechanical properties, for example almost 400 % growth was observed in elongation at break of extruded 80/20 PET/PA blends in the presence of 0.2 % compatibilizing additive compared to the sample without additive, meanwhile its strength has also improved.

**Keywords:** compatibilizing, PET, recycling, olefin-maleic-anhydride copolymer, polymer processing

## 1. Introduction

Nowadays the plastic recycling has growing importance supported by legislations in European Union where a number of aims have been accepted such as collection of plastic bottles has to reach 90 % by 2029 as well as 25 % of the material of bottles must be recycled material by 2025 and 30 % by 2030[1]. Consequently the waste bottles are necessary to be recycled. It can be solved by mechanical recycling but the presence of thermal and hydrolytic degradation generally cause significant impediment during processing. The polar plastic, PET is capable of absorbing the moisture from the air, therefore conditioning up to 0.02 % moisture

contents highly required[2]. PET with other thermoplastics (PC, ABS and PA) indicate limited compatibility and weak interfacial interaction between the components supported by SEM images (Fig 1-2).



Figure 1. SEM image of 60/40 PET/PC

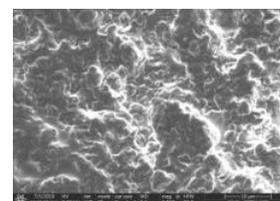


Figure 2. SEM image of 80/20 PET/PA

## 2. Experimental

In our research a real industrial waste based PET from selective collection and its blends with three engineering plastics (ABS, PC and PA) were investigated. The waste based PET were analyzed by FT-IR spectroscopy indicating polyolefin impurities coming from cap and label of PET bottles. The engineering thermoplastics were commercial, ABS (POLYLAC PA-737, density (23 °C): 1.04 g/cm<sup>3</sup>; MFI (200 °C, 5 kg): 2.6 g/10 min), PC (PANLITE L-1225L, density(23 °C): 1.20 g/cm<sup>3</sup>; MVR (300 °C, 1.2 kg):18 cm<sup>3</sup>/10 min) and PA (DOMAMID 6AV, density(23 °C): 1.14 g/cm<sup>3</sup>; MVR (275 °C, 5 kg): 165 cm<sup>3</sup>/10 min).

As compatibilizing additives four distinct types of additives have been applied, one of them was the commercial maleic-anhydride grafted polypropylene (LICOMONT AR504) supplied by Clariant GmbH, the other three additives were experimental olefin-maleic-anhydride copolymer based types, which have been used in the form of masterbatches.

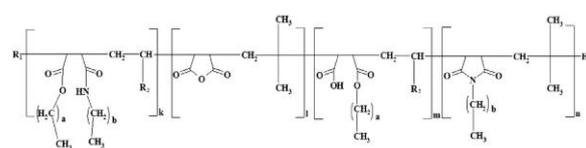


Figure 3. Structure of experimental olefin-maleic-anhydride copolymer based additives

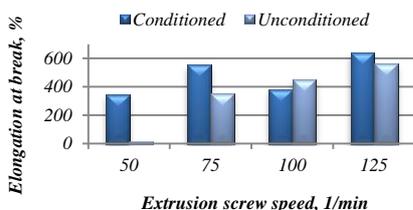
(R<sub>1</sub>): alkyl group with length of the olefinic monomer with R<sub>1</sub>-2 carbon number; a: 3-40; b: 3-32; k: 0.2-2; l: 1-7; m: 1-7 and n: 0.3-2)

Matrix materials of masterbatches were polyolefins, two different types of polypropylene (MOPLEN EP300K; density (23 °C): 0.9 g/cm<sup>3</sup>; MFR (230 °C, 2.16 kg):4.0 g/10 min, and BRASKEM H734-52; density (23 °C): 0.9 g/cm<sup>3</sup>; MFR (230 °C, 2.16 kg): 52 g/10 min) and a HDPE (HOSTALEN GC7260; density (23 °C): 0.960 g/cm<sup>3</sup>; MFR (190 °C, 2,16 kg): 8.0 g/10 min).

To achieve proper homogeneity of the samples, engineering thermoplastics were extrusion moulded by a laboratory twin screw extruder (LABTECH Engineering Company LTD LTE 20-44) adjusted with 125 1/min screw speed. Moreover blends were injection moulded by a laboratory injection moulding machine (TOPFINE A50). Before processing, raw materials were conditioned at least for 16 hours at 60 °C in a drying oven to avoid hydrolytic degradation. Masterbatches were produced by a two-roll mill (LabTech Engineering Ltd., LRM-100) at 140 °C-180 °C.

### 3. Results

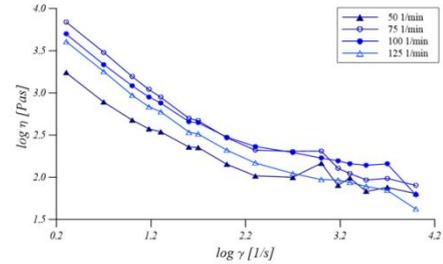
First of all investigation of the effects of thermal and hydrolytic degradation of waste based PET was required because of their negative consequence on the mechanical properties. Thus, we have studied the effects of extrusion speed (50, 75, 100, 125 1/min) as the residence time in extruder denotes to the rate of thermal degradation. Furthermore effects of the moisture content of raw material before processing was also examined thus defining the rate of hydrolytic degradation. Mechanical properties were measured by an INSTRON 3345 universal tensile testing machine with 90 mm/min crosshead speed at RH of 55% and 21 °C and the rheological behaviour of samples were executed by a capillary rheometer (CEAST Smart RHEO 2000).



**Figure 4.** Effects of conditioning and screw speed on the elongation at break in PET samples

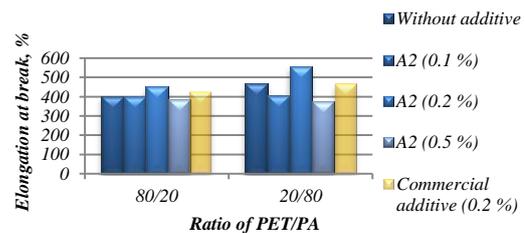
Figure 4. demonstrates elongation at break of PET samples extruded with different parameters and moisture content before processing. Both factors had effects on degradation, sample produced at 50 1/min extrusion screw speed from raw material without conditioning had the largest rate of degradation. In contrast, the sample produced with 125 1/min showed the lowest level of deterioration. Comparing samples from unconditioned raw materials with samples from conditioned one, moisture content had significant negative effects in almost all cases. Consequently, the results from tensile tests reveal that sample processed at the highest extrusion screw speed has the highest elongation at break and tensile strength irrespectively of pre-treatment of raw material. The rheological behaviour of samples confirmed the results of tensile tests, namely the most stable curve was observed in case

of the sample produced with 125 1/min screw speed (Fig. 5.)



**Figure 5.** Viscosity-shear rate of PET samples with different screw speed

Blends both with 80 % and 20 % PET content showed improved mechanical properties in the presence of olefin-maleic-anhydride copolymer based additive having more nitrogen containing functional groups compared to the other additive structures. We have investigated three additive concentrations of that additive structure the most advantageous was 0.2 %. Tensile strength gained by about 2 MPa while in case of in elongation at break almost 50 % and more than 90 % growth of sample with 80 % PET content and of sample with 20 % PET content were measured, respectively.



**Figure 6.** Effects of olefin-maleic-anhydride copolymer based additives on PET samples with different concentrations

The matrix type of masterbatches were decided, while using the mentioned additive with other type of matrix in 80/20 PET/PA indicated approximately 395 % increase in elongation at break while the tensile strength was the same compared to the sample without additive.

### 4. Conclusion

The inference can be drawn from our experiment that the largest extrusion screw speed should be applied during processing of PET and the conditioning of raw material is also required to achieve good mechanical properties. The limited compatibility between the components of PET/engineering plastic blends could be improved in the presence of masterbatches from the olefin-maleic-anhydride copolymer based additives. As a reference commercial additive was also examined. We managed to produce additive structures behaving in a successful way in blends both of higher PET and of higher PA content.

### References

- [1] Polymers, June 2019. Vol. 5 No. 6, 549.
- [2] Firas Awaja, Dumitru Pavel: Recycling of PET, European Polymer Journal 41, 1453-1477, 2005