

# MFC Concept as a Possible Solution for Closed-loop Recycling of Food Packaging Trays

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**Abstract.** Flexible plastic films from food packaging trays make up one of the largest fractions of the plastic waste stream and recycling is one of the most important actions to deal with this fraction, reducing the impact of these plastics on the environment. However, the recyclability of multi-layered films is not straightforward and in most of the cases these fractions are landfilled or incinerated. Recycling of discarded mixed polymers without previous separation often results in low mechanical properties which have lead researchers to investigate novel solutions for recycling. In this research, the concept of microfibrillar composites (MFCs) was investigated aiming to upcycle mixed polymer waste streams. A blend based on polypropylene (PP) and poly(ethylene terephthalate) (PET) at a weight ratio of 80/20 PP/PET was studied. The final step of the MFC processing was conducted using a conical twin screw extrusion. The morphological results confirmed the presence of PET microfibrils in the composites, leading to an improvement in mechanical properties such as in the tensile yield strength and strain at break. Subsequently, the MFC samples were successfully moulded into trays via thermoforming.

**Keywords:** microfibrillar composites, morphology, mechanical properties, recycling

## INTRODUCTION

Polymer products became an integral part of the modern life. With the increase in plastics consumption, the biggest challenge is how to solve the issue of plastic waste disposal, lowering its impact on the environment [1]. The success of polymers is mainly attributed to their low cost, low density and numerous mechanical and thermal properties that make them suitable for a wide range of applications. Packaging materials such as food packaging trays typically have very short lifetimes and a huge amount of plastic waste is generated from packaging applications. The most used polymers in food packaging applications are polyolefins and polyesters, and quite often these products are produced in complex multi-layered films, hindering the separation of the individual polymers in those plastic products. It is well known that many polymers are immiscible in the melt state, which causes phases separation and decreasing mechanical properties of the final mechanically recycled product [2]. Therefore, many researchers [3-5] investigated new methods to overcome the problem of immiscibility in polymer waste streams aiming to improve the processing of recycled blends and the final properties of the materials. The most common approach to mitigate the immiscibility is the addition of compatibilizers or different types of additives into polymer mixtures [2, 6]. Alternatively, production of microfibrillar composites (MFC) was presented as a worthy and easy way of making new composites with increased mechanical properties [7, 8]. The key factor in the development of MFCs lies in the immiscibility of two polymers, as this is one of the important requirements for their production, as well as the difference in melting temperatures (e.g. between polyolefins and polyesters).

Hence, considering the previous studies and proposals, this research focuses on developing the MFC concept as a possible solution for the recycling of food packaging trays. The samples were prepared in a similar way to the conventional MFC production, except for the last production step, where extrusion was used instead of injection moulding. Until now, there are no known studies where extrusion and thermoforming were used as final steps in the MFC processing. Therefore, virgin polymers were used in this study due to possible limitations that might be encountered during processing of recycled mixtures, such as degradation, viscosity issues, contaminations etc. [5, 9]. Our goal is to produce the MFC sample into a sheet via extrusion and convert it into a thermoformed tray. In our previous work, the influence of compatibilizers was pointed out, as well as the improvement of the mechanical properties such as tensile yield strength and strain at break obtained with rubber-based compatibilizers [4]. During the thermoforming process, the composite is exposed to extensive stretching; hence, the strain at break is an important characteristic. Therefore, a compatibilizer was also used in this study. The presence of the PET microfibrils in the

composites may contribute to increased mechanical properties and possibly decrease permeability. Therefore, MFCs can be a possible solution for the multi-layered packaging waste, enabling a completely closed-loop recycling, however, at this point without taking into account the food contact legislation.

## EXPERIMENTAL

### Materials

The virgin materials used in this study were polypropylene (PP) purchased from Sabic (Sabic 575P, Bergen op Zoom, The Netherlands) with a melt flow rate (MFR) of 11 g/10 min (2.16 kg, 230 °C), and poly(ethylene terephthalate) (PET) (LIGHTER C93) from Equipolymers (Schkopau, Germany), which is a bottle-grade material with an intrinsic viscosity of  $0.80 \pm 0.02$  dL/g. PET was dried in a vacuum oven for 15 h at 80 °C, and 2 h before processing at 120 °C, while PP was used as received. Polyolefin-based elastomer grafted with maleic anhydride (POE-g-MA), (Acti-Tech 16MA13) was used as a compatibilizer in this study, which is an Exxon performance polymer based, kindly donated by Nordic Grafting Company (NGC, Hellerup, Denmark). The grafting percentage of the maleic anhydride (MA) group onto the backbone of the compatibilizer was 1.3 wt%, according to the data sheet. Compatibilizer was dried at 60 °C for 15 h before processing.

### Sample Preparation

The samples were prepared in a weight ratio of 80/20 PP/PET, where PP was used as a matrix and PET as a reinforcing component. To compatibilize the PP/PET blend, POE-g-MA was added in 6 wt %, while the same PP/PET ratio was maintained. The preparation of the blends (with and without compatibilizer) was conducted via twin screw extruder (Coperion ZSK18, Stuttgart, Germany) with two co-rotating screws of 18 mm diameter,  $L/D = 40$  and a die opening of 19 mm  $\times$  2 mm. The screw speed was set at 120 rpm and the barrel temperatures were set between 205 and 260 °C. The extrudate was obtained as a sheet with dimensions of 25 mm  $\times$  1 mm, by passing through calender rolls, which were cooled down to  $\sim 15$  °C. The received cooled extrudate was cold drawn by a pair of rolls passing through a hot oven (200 °C, 55.5 cm  $\times$  60 cm) above the glass transition temperature of PET. During drawing, the surface temperature of the extrudate was measured and amounted to approximately 95 °C. The speed of the rolls was adjusted to obtain a draw ratio of 8. Shredded pure blend and microfibrillar blends were extruded again via conical twin screw extruder (MAS24, Schulz GmbH, Hobelweg, Austria) into sheets with dimensions 150 mm  $\times$  1 mm. The conical twin screw extruder was set at temperatures of 170, 190, 200 and 210 °C and speed of 70 rpm. The obtained sheets were cooled down by passing between cast rolls at a temperature of 60 °C. The MFC sheets were thermoformed by a Formech508FS machine (Harpeden, United Kingdom) at 135 °C for 65-75 seconds into round trays with a diameter of 55 mm and 10 mm height.

### Characterization

Small angle light scattering (SALS) was used to determine the spherulites size of the PP in the final sheets. The samples were microtomed into 15  $\mu$ m thick layers and placed between two microscopic slides. SALS patterns were obtained with a 632.8 nm He-Ne laser (1 mm beam size) with the source of polarized monochromatic light. SALS  $H_v$  patterns were captured using a Hamamatsu digital camera (Hamamatsu Photonics K.K., Hamamatsu City, Japan) and analysed with Hipic 6.3.0 software (Hamamatsu Photonics K.K., Hamamatsu City, Japan). The equivalent radius ( $R_0$ ) of the spherulites was estimated with the following Equation (1):

$$R_0 = \frac{1025}{\pi} \frac{\lambda}{\sin\left(\frac{\theta_{max}}{2}\right)},$$

where  $\lambda$  is the wavelength of light in the medium. The distance from the centre of the  $H_v$  pattern to the intensity maximum in one lobe, in conjugation with the known sample-to-film distance, is a measure of the polar angle  $\theta_{max}$ . ( $\theta_{max} = \tan(\text{distance from the centre of the } H_v \text{ pattern to the intensity maximum in one lobe/sample-to-film distance})$ ) [4]. The structural characterization of the samples was performed by scanning electron microscopy (SEM) FEG SEM JEOL JSM-7600F 202 (Tokyo, Japan). The sheets were immersed in liquid nitrogen and subsequently fractured, and afterwards the surfaces were sputtered with gold by a Bal-Tec SCD005 sputter coater (Bal-Tec, Balzers, Liechtenstein). Micrographs were obtained with an accelerating voltage of 15 kV. Raman spectroscopy was used as an additional

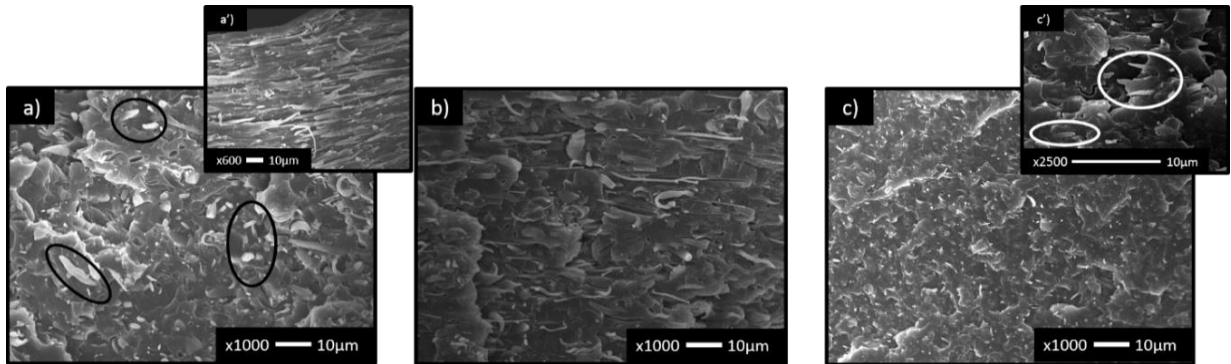
technique to confirm the dispersion of PET fibrils in the composites. Raman spectra were obtained on a Horiba LabRAM HR Evolution confocal microscope (Horiba Scientific, Longjumeau, France) using a laser excitation of 785 nm. The objective lens ( $\times 100$ ) was used to focus the laser onto the sample. The scanned Raman red maps  $40 \times 35 \mu\text{m}$  were obtained with a step size of  $1 \mu\text{m}$  and analysed using the LabSpec 6 software (also from Horiba Scientific).

The tensile bars (ASTM D412T.C) were die cut from the sheets with dimensions  $115 \text{ mm} \times 6 \text{ mm} \times 1 \text{ mm}$  and gauge length of 33 mm. They were tested by an Instron 5565 tensile device (Norwood, MA, USA) according to standard ISO 527. During the tests, two test speeds were used: 1 mm/min as a first cross-head rate, and 5 mm/min after the Instron dynamic extensometer was removed (type catalogue 2620-603 with a gauge length of 12.5 mm). Analysis of the results was performed with the Bluehill software. The differences between the samples were calculated by t-independent test via the software package SPSS Statistics 24 (Armonk, NY, USA) with a probability value of 0.05.

## RESULTS AND DISCUSSION

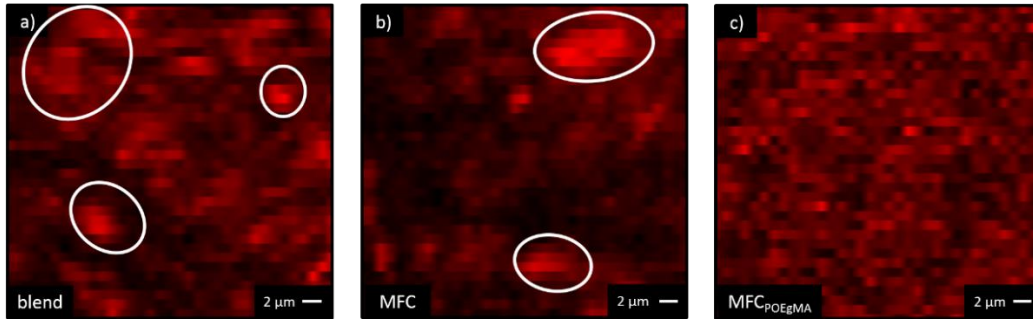
### Morphology study

It is well known that the structural development is of huge importance for good mechanical properties of composites [4], hence the extruded sheets were subjected to intensive microscopy study via SEM and Raman spectroscopy. In Figure 1, SEM micrographs of the blend, MFC and MFC<sub>POEgMA</sub> sheets in longitudinal direction are presented. As can be seen, the blend shows a non-uniform dispersion and distribution of PET particles with a coalescence phenomenon present (black circles in Figure 1a) in the centre of the sheets, while at the edges of the sheet (Figure 1a'), the presence of stretched PET particles can be noticed. These stretched particles could be a consequence of the extrusion process as the final processing step and high viscosity of the melt, which has made the production of the sheets more difficult. We assumed that beside the high viscosity, another reason could be increased shear rate in die edges; hence, an additional stretching may have happened.



**FIGURE 1.** Microstructures of the samples obtained via SEM in longitudinal direction: (a), (a') blend, (b) MFC, (c), (c') MFC<sub>POEgMA</sub>

Further on, in the MFC sample PET microfibrils were detected with high aspect ratios (Figure 1b), as well as a good level of orientation. Although, some fibril clusters were also found along the MFC sample. The reason for the clusters formation might be the relaxation of PET fibrils that takes place during the extrusion process. Observing the MFC containing POE-g-MA, it can be noticed that the fibrils are much shorter (Figure 1c') than in the non-compatible MFC, however, a relatively uniform dispersion and distribution can be noticed (Figure 1c). The presence of the compatibilizer may prevent coalescence of the fibrils by covering the PET particles during melt blending, which facilitates the dispersion of droplets and consequently reducing the final fibril lengths [4]. The red maps obtained via Raman spectroscopy, shown in Figure 2, confirm the dispersion of the second phase in the matrix. The maps show PP as a black phase, while the red one represents the PET phase. The red phase in Figures 2a and 2b show the PET clusters which have been detected in the composites while in the case of MFC<sub>POEgMA</sub> it can not be seen. Similar findings have been reported in our previous study [4].



**FIGURE 2.** Red maps of the samples obtained via Raman spectroscopy: (a) blend, (b) MFC, (c) MFC<sub>POEGMA</sub>. Matrix PP represents black phase, and PET is the red phase.

In addition to the structural development, the average spherulite size of PP was quantitatively measured by SALS. The average diameters of the PP spherulites in the composites are listed in Table 1. The MFC<sub>POEGMA</sub> shows the lowest spherulite size when compared to the blend and the MFC. The reason for that lies in well dispersed and distributed PET fibrils which have a strong nucleating effect on the PP matrix, since they are covered by the compatibilizer [4]. In our previous work [4], it was explained that the nucleating effect may arise from isolated compatibilizer particles too, as there is always a certain amount of compatibilizer that does not react with the PET.

**TABLE 1.** Average diameter of PP spherulites in the composites

Material	Spherulite diameter, $\mu\text{m}$
Blend	$10.4 \pm 1.0$
MFC	$8.3 \pm 0.9$
MFC <sub>POEGMA</sub>	$3.6 \pm 0.7$

## Tensile Properties

In order to discuss mechanical properties, the tensile modulus, tensile yield strength of the samples and strain at break were defined and the results are listed in Table 2. An increase in stiffness was observed for the MFC sample in comparison with the blend and the MFC<sub>POEGMA</sub>, which can be explained by the presence of long PET fibrils which will contribute to increased stiffness. Due to the presence of the long fibrils in this sample, it is considered that sufficient interfacial contact between the microfibrils and the matrix phase is present, even without the presence of a compatibilizer [4]. MFC<sub>POEGMA</sub> shows a lower stiffness, due to the shorter PET fibrils caused by the presence of the compatibilizer containing a rubber (soft) backbone [2, 4] and higher strength in comparison to other two samples. The reason for higher tensile yield strength for MFC<sub>POEGMA</sub> lies in a better adhesion between the phases. Both PP and PET uniformly stretch during the tensile test. Furthermore, MFC<sub>POEGMA</sub> shows a higher strain at break result, which confirm that stress transfer between the fibrils and the matrix is considerably improved.

On the other hand, the blend presented a higher stiffness, probably due to the presence of the PET fibrils at the edges of the sample (see Figure 1a'), but they did not affect the tensile yield strength. The stress transfer in the blend is not enough effective due to lower adhesion between phases, which causes for the lower yield strength value, as well as the low strain at break.

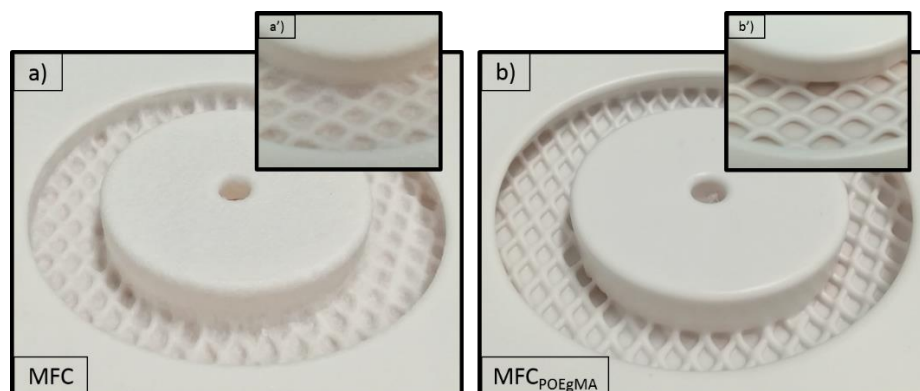
**TABLE 2.** Tensile properties of extruded MFC plates

Material	Tensile Modulus, GPa	Yield Strength at Zero Slope, MPa	Strain at break, %
Blend	$1.91 \pm 0.09$	$28.6 \pm 0.5$	$15.9 \pm 1.2$
MFC	$2.15 \pm 0.13$	$30.3 \pm 0.9$	$14.5 \pm 3.8$
MFC <sub>POEGMA</sub>	$1.59 \pm 0.30$	$32.5 \pm 5.5$	$250.0 \pm 229.0$

## Preliminary Thermoform Experiments

As shown from the results above, there is an improvement in mechanical properties of the MFC samples, particularly in the yield strength and the strain at break for the sample MFC<sub>POEGMA</sub>. In Figure 3, thermoformed parts are shown for the samples MFC and MFC<sub>POEGMA</sub>. As can be seen from the images, MFC<sub>POEGMA</sub> shows a smooth and

uniform surface, while the MFC sample shows a rougher surface. From Figures 3a' and 3b', can be noticed that details in the MFC<sub>POEgMA</sub> are more profound than in the MFC sample. Comparing the strains at break values, it is clear that MFC<sub>POEgMA</sub> has a higher ability to stretch during the thermoforming process and thus obtained a better shape at the edges then the MFC sample. Moreover, during the thermoforming process, the MFC sample would break much easier due to its brittleness.



**FIGURE 3.** Thermoformed parts for the MFC samples: (a), (a') MFC, (b), (b') MFC<sub>POEgMA</sub>.

## CONCLUSION

This research presented the MFC concept as a possible solution for the recycling of food packaging trays aiming at a closed-loop recycling path.

Microstructural analysis has confirmed the presence of PET microfibrils in the MFC samples, as well as an improved dispersion and distribution in the MFC<sub>POEgMA</sub> sample. SALS measurements showed small spherulite sizes in all samples, confirming the theory of heterogeneous nucleation of PP promoted by the PET fibrils.

The mechanical results showed a significant increase in the tensile yield strength and in the strain at break for the sample MFC<sub>POEgMA</sub>, as the compatibilizer promoted a better interfacial adhesion between PP and PET phases. Based on these findings, the MFC samples were successfully moulded into trays via thermoforming.

In the future, the optimisation of the thermoforming process will be necessary, likewise taking into consideration the food contact regulations as the products would be prepared from recycled mixtures. Moreover, investigation of the permeability of the trays and its relation to deformation of PET fibrils during thermoforming would be of high importance.

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